

Program of the **MDB 2026**

Material Development for Batteries (MDB):

Progresses and Challenges

Sep. 28 – Oct. 2, 2026

Sejong University, Seoul, South Korea

Organizing Committee

- Conference Co-Chair

- Prof. Dr. Payam Kaghazchi, Forschungszentrum Juelich GmbH, Germany; University of Twente, The Netherlands
- Prof. Seung-Taek Myung, Sejong University, South Korea

- Scientific Committee

- Prof. Dr. Payam Kaghazchi, Forschungszentrum Jülich GmbH, Germany
- Prof. Seung-Taek Myung, Sejong University, South Korea
- Prof. Yang Zhao, University of Western Ontario, Canada
- Prof. Javier Carrasco, CIC energiGUNE, Spain
- Prof. Yongcheng Jin, Ocean University of China, China
- Prof. Tzu-Ho Wu, NTUST, Taiwan
- Prof. Jinkwang Hwang, Kyoto University, Japan
- Prof. Giuseppe Antonio Elia, Politecnico di Torino, Italy

Scientific Program

- Alkali-ion batteries (Li-, Na, and K-)
- Alkali metal batteries (Li-, Na, and K-)
- Solid-State-Batteries
- Simulation, Modeling, and Machine learning/AI

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MDB 2026 Material Development for Batteries (MDB): Progresses and Challenges

Conference Schedule Sep.28 – Oct. 2, 2026 Sejong University, Seoul, South Korea

	Monday 28 September	Tuesday 29 September	Wednesday 30 September	Thursday 1 October	Friday 2 Oct.
Morning 1	Chair: Prof. Payam Kaghazchi	Chair: Prof. Natalia Voronina	Chair: Prof. Jinkwang Hwang	Chair: Prof. Tzu-Ho Wu and Prof. Meng-Chang Lin	Open discussion & networking
	Coffee Break				
Morning 2	Chair: Prof. Payam Kaghazchi	Chair: Prof. Shu-Hao Chang	Chair: Prof. Natalia Voronina	Chair: Prof. Giuseppe Antonio Elia	
	Lunch				
Afternoon 1	Chair: Prof. Heng Zhang and Prof. Jeongsik Yun	Chair: Prof. Claudio Cazorla and Prof. Aishuak Konarov	Chair: Prof. Shinichi Kumakura and Prof. Aishuak Konarov	Chair: Prof. Yongcheng Jin and Prof. Payam Kaghazchi	
	Coffee Break				
Afternoon 2	Chair: Prof. Payam Kaghazchi and Prof. Kingo Ariyoshi	Chair: Prof. Payam Kaghazchi and Prof. Sheng-Heng Chung	Chair: Prof. Yauhen Aniskevich and Prof. Hiroaki Kobayashi	Chair: Prof. Jiefang Zhu and Prof. Shuai Yuan	
	Dinner		Banquet	Dinner	

Monday 28 September 2026 – Morning

09:00 – 9:30 **Registration**

09:30 – 9:45 **Opening**

Room: 1

Chaired by Payam Kaghazchi

09:45 – 10:25 **Plenary**

Bing Joe Hwang (National Taiwan University of Science and Technology, Taiwan)

Decoding and Controlling Interfacial Dynamics for Next-Generation Lithium–Sulfur Batteries

10:25 – 10:45

Coffee Break

10:45 – 11:15 **Keynote**

Sang-Young Lee (Yonsei University, South Korea)

Low-Pressure, Low-Cost All-Solid-State Batteries Enabled by Monomer-in-Salt Electrolytes

11:15 – 11:45 **Keynote**

Fu-Ming Wang (National Taiwan University of Science and Technology, Taiwan)

How to improve the performance of $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$: the point of view from crystal structure to electrolyte additive

11:45 – 12:05 **Invited**

Shinichi Kumakura (Tokyo University of Science, Japan)

Impact of Composition and Morphology on the Electrochemical Performance of Li-Rich Layered Oxides

12:05 – 12:25 **Invited**

Jeongsik Yun (Incheon National University, South Korea)

Formation Mechanism of Radially Oriented
Microstructures in Tungsten-Doped Ni-Rich Layered
Oxides Cathodes

12:30 – 13:30

Lunch

Monday 28 September 2026 – Afternoon

Room: 1

Chaired by Heng Zhang

13:30 – 13:50 **Invited**

Huanan Duan (Shanghai Jiao Tong University, China)

Synergistic Modulation of Interfacial Chemistry and
Residual Solvent Chemistry in Composite Solid
Electrolytes

13:50 – 14:10 **Invited**

Chia-Chin Chen (National Taiwan University, Taiwan)

Visualizing and Unraveling Charge Transport in Battery
Electrodes

14:10 – 14:30 **Invited**

Hyungsub Kim (Korea Atomic Energy Research Institute,
South Korea)

Controlling Lithiation Pathways for Low-Defect Ni-
Rich Layered Cathodes

Chaired by Jeongsik Yun

14:30 – 14:50 **Invited**

Heng Zhang (Huazhong University of Science and Technology,
China)

Sulfonimides as Negative Charges for Advanced Battery
Electrolytes

14:50 – 15:10 **Invited**

Wei-Ren Liu (Chung Yuan Christian University, Taiwan)

Oxygen vacancies engineering to boost Li-ion transport
and cycling stability in TiNb_2O_7 anodes

15:10 – 15:30 **Invited**

Kingo Ariyoshi (Osaka Metropolitan University, Japan)

Asymmetric Li Insertion/Extraction

Kinetics of $\text{Li}[\text{Li}_{1/3}\text{Ti}_{5/3}]\text{O}_4$

15:30 – 15:50

Coffee Break

Chaired by Payam Kaghazchi

15:50 – 16:30 **Plenary**

Xueliang Andy Sun (Eastern Institute of Technology, China)

Halide-based Electrolytes for All Solid-State Batteries

16:30 – 16:50 **Invited**

Richard Fields (Cellerate Ltd, United Kingdom)

Precision Assembled Single Layer Pouch Cells with
Reference Electrode for Studying Emerging Battery
Chemistries

16:50 – 17:10 **Invited**

Moulay Tahar Sougrati (Politecnico di Torino, France)

Direct Regeneration of Olivine Cathodes: LFP Successes
and Emerging Challenges with LMFP and Blends

17:10 – 17:30 **Invited**

Matteo Bonomo (Sapienza University of Rome, Italy)

Reinventing Electrolytes: Deep Eutectic Solvents for
Energy Storage

Chaired by Kingo Ariyoshi

17:30 – 17:50 **Invited**

Chao Zhang (Uppsala University, Sweden)

AI-Accelerated Modelling and Design of Liquid Electrolyte
for Rechargeable Batteries

17:50 – 18:10 **Invited**

Di Wang (Kyoto University, Japan)

Assessment of Thin Lithium Metal Electrodes via Voltage
Profile Diagnostics

18:10 – 18:25 **General**

Jaehyun Park (Korea Electrotechnology Research Institute,
South Korea)

Micron-scale Fe_2P Modified with an N-doped Carbon
Layer for Efficient Polysulfide Regulation and Sulfur
Redox Conversion

18:30 –

Dinner

Tuesday 29 September 2026 – Morning

Room: 1

Chaired by Natalia Voronina

09:00 – 09:15 **General**

Zhang Hao (Harbin Institute of Technology, China)

Brief Ball Milling Coupled with Low-Dose Molten Salt
Recrystallization: Synchronous Particle Homogenization
and Intrinsic Al Impurity In-situ Doping toward High-
Voltage Stable Recycled LiCoO₂

09:15 – 09:30 **General**

Manojkumar Seenivasan (Ming Chi University of
Technology, Taiwan)

Toward Co-Free Single-Crystal Ni-Rich Cathodes:
Challenges and Progress

09:30 – 10:00 **Keynote**

Zhumabay Bakenov (Nazarbayev University, Kazakhstan)

Pilot-Scale Prototyping and Validation of Lithium-
Ion Pouch Cells for Electric Vehicle Applications

10:00 – 10:20 **Invited**

Shu-Hao Chang (Chung Yuan Christian University, Taiwan)

Current Collectors and Interfacial Engineering for
Anode-Free Lithium Metal Batteries

10:20 – 10:40 **Invited**

Miguel Ángel Muñoz-Márquez (International Iberian
Nanotechnology Laboratory, Portugal)

From Native Lithium Surface Chemistry to
Temperature-Dependent SEI Evolution in Lithium-Metal
Batteries

10:40 – 11:00

Coffee Break

Chaired by Shu-Hao Chang

11:00 – 11:20 **Invited**

Jiefang Zhu (Uppsala University, Sweden)

Interfacial Engineering of Lithium Metal for High-Energy Batteries: Insights from Redox-Mediator-Based Li-O₂ Systems

11:20 – 11:40 **Invited**

Shih-kang Lin (National Cheng Kung University, Taiwan)

Microstructure and electrochemical performance of Li-Na-Mg alloy anode for Li metal batteries

11:40 – 12:00 **Invited**

Kazuki Yoshii (National Institute of Advanced Industrial Science and Technology, Japan)

Charge-Discharge Behavior of Li-Mg Alloy Electrodes in Ionic Liquid Electrolytes

12:00 – 12:20 **Invited**

Koji Yazawa (JEOL ltd, Japan)

Operando NMR spectroscopy for battery materials

12:30 – 13:30

Lunch

Tuesday 29 September 2026 – Afternoon

Room: 1

Chaired by Claudio Cazorla

13:30 – 13:50 **Invited**

Oier Arcelus (CIC EnergiGUNE, Spain)

Phase-Field Modelling of Microstructural Evolution and Degradation in Battery Materials

13:50 – 14:10

Break

14:10 – 14:30 **Invited**

Kourosh Malek (Forschungszentrum Jülich GmbH, Germany)

Developing a Cloud-Connected Platform for Energy Materials Acceleration

Chaired by Aishuak Konarov

14:30 – 14:50 **Invited**

Guowei ZHAO (Huanggang Normal University, China)

Synthesis and Characterization Technologies for Li_2S as a Key Raw Material in Sulfide-type Solid-State Electrolytes

14:50 – 15:10 **Invited**

Yaoyu Ren (Tsinghua University, China)

AI-aided Development of Low-Cost Halide Solid Electrolytes

15:10 – 15:30 **Invited**

Claudio Cazorla (Universitat Politècnica de Catalunya, Spain)

Universal Insights into Ion Transport in Solid- State Electrolytes

15:30 – 15:50

Coffee Break

Chaired by Payam Kaghazchi

15:50 – 16:30 **Plenary**

Shinichi Komaba (Tokyo University of Science, Japan)

Exploring Polyanionic Compounds for K-Ion Batteries

16:30 – 16:50 **Invited**

Jiangwei Ju (Qingdao Institute of Bioenergy and Bioprocess Technology, China)

Tuning carrier transport for fast charging sulfide based all-solid-state batteries

16:50 – 17:10 **Invited**

Sheng-Heng Chung (National Cheng Kung University, Tainan)

Functional lithium halide coating layer for thin- lithium-sulfur cells

17:10 – 17:30 **Invited**

Eric Jianfeng Cheng (Tohoku University, Japan)

Garnet-solid electrolytes: processing, interfaces, and prospects for solid-state lithium-metal batteries

Chaired by Sheng-Heng Chung

17:30 – 17:50 **Invited**

Mauricio Rincon Bonilla (Basque Center for Applied Mathematics, Spain)

Mitigating Aluminium Contamination in Garnet Solid Electrolytes via Rational Substrate Design

17:50 – 18:10 **Invited**

Yu-Sheng Su (National Yang Ming Chiao Tung University, Taiwan)

Interfacial Regulation of Lithium Metal Anodes via
Nitrate-Induced Lithiophilic Frameworks for Ultralow
Overpotential Alkali Metal
Batteries

18:10 – 18:30 **Invited**

Seongjae Ko (The University of Tokyo, Japan)

Beyond 99.9% Coulombic Efficiency in Aqueous Zinc
Metal Batteries Without Relying on SEI

18:30 –

Dinner

Wednesday 30 September 2026 – Morning

Room: 1

Chaired by Jinkwang Hwang

09:00 – 09:40 **Plenary**

Yamada Atsuo (The University of Tokyo, Japan)

Hydration thermodynamics of $\text{Na}_2\text{Fe}_2(\text{SO}_4)_3$ defines its ambient storage and closed-loop recycling conditions

09:40 – 10:10 **Keynote**

Linjie Zhi (China University of Petroleum, China)

Understanding the Materials and Chemistry in Rechargeable Li(Na)/Cl Batteries

10:10 – 10:40 **Keynote**

Kazuhiko Matsumoto (Kyoto University, Japan)

Behavior of Sodium Metal Electrode and Some Effective Additives

10:40 – 11:00

Coffee Break

Chaired by Natalia Voronina

11:00 – 11:20 **Invited**

Shuai Yuan (Shanghai University, China)

Polymer-based solid-state electrolytes

11:20 – 11:40 **Invited**

Hamideh Darjazi (Politecnico di Torino, Italy)

Balancing ion transport, high-voltage stability and sustainable design in solvent-free UV-crosslinked solid polymer electrolytes

11:40 – 12:00 **Invited**

Naoto Tanibata (Nagoya Institute of Technology, Japan)

Designing High-Deformability Materials for All-Solid-State Batteries Through Shear Modulus Engineering

12:00 – 12:20 **Invited**

Masaki Matsui (Hokkaido University, Japan)

Moisture-Assisted Hydroflux Synthesis of Layered Cathodes

12:30 – 13:30

Lunch

Wednesday 30 September 2026 – Afternoon

Room: 1

Chaired by Shinichi Kumakura

13:30 – 14:00 Keynote

Naoaki Yabuuchi (Yokohama National University, Japan)

Defect-engineered Oxide-based Electrode Materials for Li Storage Applications

14:00 – 14:20 Invited

Mario Marinaro (ZSW, Germany)

NFPP Cathodes for Sodium-Ion Batteries:
From Material Design to Scale-Up

14:20 – 14:40 Invited

Hiroaki Kobayashi (The University of Tokyo, Japan)

Utilization of Cationic Multi-electron Redox in Na-Superrich Oxides for Sodium-Ion Battery

Chaired by Aishuak Konarov

14:40 – 15:00 Invited

Changhee Lee (Tokyo University of Science, Japan)

Na-Layer Chemistry and Cathode Properties of P2/P3-Type Layered Oxides

15:00 – 15:20 Invited

Jongsoon Kim (Sungkyunkwan University, South Korea)

Enhancing Oxygen Redox Kinetics in Na-Layered Cathodes toward High-Power and High-Energy Sodium-Ion Batteries

15:20 – 15:40 Invited

Jinkwang Hwang (Kyoto University, Japan)

Sustainable Chemical Delithiation of LiFePO₄ and Its
Reuse for Sodium Batteries

15:40 – 16:00

Coffee Break

Chaired by Yauhen Aniskevich

16:00 – 16:20 **Invited**

Hyung-Seok Kim (Korea Institute of Science and Technology,
South Korea)

Stabilizing Oxygen Redox in Sodium Manganese Oxide
Cathodes through Doping and Cooling Engineering

16:20 – 16:40 **Invited**

Ramesh J. Deokate (Science and Commerce College, India)

Nanostructured Sodium Vanadium Phosphate-Based
Cathode Materials for Sodium-Ion Rechargeable Batteries

16:40 – 17:00 **Invited**

Aishuak Konarov (Nazarbayev University, Kazakhstan)

Optimized Hard Carbon for High-Performance
Sodium-Ion Batteries

Chaired by Hiroaki Kobayashi

17:00 – 17:20 **Invited**

Yauhen Aniskevich (Silesian University of Technology,
Poland)

Beyond Graphite: Na⁺ - Solvent Co-Intercalation in
Organic and Carbon Hosts

17:20 – 17:40 **Invited**

Tzu-Ho Wu (National Taiwan University of
Science and Technology, Taiwan)

Electrochemical activation of V³⁺ and V⁴⁺ oxides in
aqueous zinc-ion batteries

17:40 – 18:00 **Invited**

Fan-Wei Liu (National Chung Hsing University, Taiwan)

Recycling, Recovery, and Reuse of High-Purity V2O5
Cathode Materials from Waste Aqueous Zinc-Ion
Batteries

18:00 – 18:20 **Invited**

Jeng-Yu Lin (Tunghai University, Taiwan)

Interfacial Engineering of MnO₂ Cathodes via MOF- 5
Thin-Film Modification for Ultra-stable Aqueous Zinc-Ion
Batteries

18:30 –

Dinner

Thursday 1 October 2026 – Morning

Room: 1

Chaired by Tzu-Ho Wu

09:00 – 09:20 **Invited**

Mark Huijben (University of Twente, Netherlands)

Fast Lithium-ion Intercalation in a Mutual Crystalline-Amorphous Single Phase

09:20 – 09:40 **Invited**

Giuseppe Antonio Elia (Politecnico di Torino, Italy)

Addressing key challenges in the development of Zn-based electrochemical storage systems

09:40 – 10:00 **Invited**

Meng-Chang Lin (National Chung Hsing University, Taiwan)

Multiple Pathways for Aluminum-Based Energy Storage across Chemistries and Scales

Chaired by Meng-Chang Lin

10:00 – 10:20 **Invited**

Ryoichi Tatara (Yokohama National University, Japan)

Rubidium Superoxide as a Stable Discharge Product for Metal-O₂ Batteries

10:20 – 10:40 **Invited**

Chaoxia Wang (Jiangnan University, China)

A Gesture Recognition Smart Glove Using Fabric-Based Conductive Carbon Spheres: Self-Powered Zinc-Air Battery System and Its Sensing Performance

11:40 – 11:00 **Invited**

Xin Su (Harbin Institute of Technology Weihai, China)

High-Energy Lithium Batteries: Synergistic Materials Design and Closed-Loop Recycling

11:00 – 11:20

Coffee Break

Chaired by Giuseppe Antonio Elia

11:20 – 11:40 **Invited**

Manuel Souto (Universidade de Santiago de Compostela, Spain)

Boosting the performance of organic cathode materials based on redox-active covalent organic frameworks for metal-ion batteries

11:40 – 12:10 **Keynote**

Dominic Bresser (Helmholtz Institute Ulm, Germany)

Critical Role of the Current Collector for Lithium-Metal Electrodes

12:10 – 12:25 **General**

B.P. Shivamurthy (Gyeongsang National University, South Korea)

Engineering fibril networks and microstructure evolution in semi-dry LFP electrodes via extrusion-assisted processing

12:30 – 13:30

Lunch

Thursday 1 October 2026 – Afternoon

Room: 1

Chaired by Yongcheng Jin

13:30 – 13:45 **General**

Hyeon Woo Choi (Gyeongsang National University, South Korea)

Development of a Continuous High-Speed Conductive Carbon Coating Process for NCM Cathode Active Materials with a Continuous Pre-Mixing

13:45 – 14:00 **General**

A. Patriarchi (University of Camerino, Italy)

Unravelling the Role of Doping in Prussian Blue Frameworks for Alkali-based Batteries

14:00 – 14:15 **General**

Bibin Jose (Forschungszentrum Jülich GmbH, Germany)

Atomistic Investigation of Fe-Substituted Mixed Polyanionic Cathodes for Sodium-Ion Batteries

14:15 – 14:30 **General**

Purna Chandra Rath (National Yang Ming Chiao Tung University, Taiwan)

A Scalable Thermal Prelithiation Strategy Using Air-Stable Lithium Precursors for High-Performance Silicon Anodes

Chaired by Payam Kaghazchi

14:30 – 14:45 **General**

Leonardo Balducci (Politecnico di Torino, Italy)

Novel Additive for Addressing Interfacial Instability in
PVDF-HFP/Pyr14FSI Systems for Lithium Metal Batteries
at Room Temperature

14:45 – 15:00 **General**

Nanasaheb M. Shinde (Ajou University, South Korea)

Boosting Electrochemical Performance of $\text{Ti}_3\text{C}_2\text{T}_x\text{@Ni}$
MXene-Based Composite Electrodes

15:00 – 15:15 **General**

Sompalli Kishore Babu (Chung Yuan Christian University,
Tiwan)

Rational Design of $\text{MnFe}_2\text{O}_4\text{@Ti}_3\text{C}_2\text{T}_x$ MXene
Heterostructures from Bimetallic MOF for High-Capacity
Li/Na-ion Batteries

15:15 – 15:30 **General**

Jianing Li (Harbin Institute of Technology, China)

Additive-Driven Regulating Solvation Structure Toward
Stable High-Voltage Lithium Metal
Batteries

15:30 – 15:50

Coffee Break

Chaired by Jiefang Zhu

15:50 – 16:05 **General**

Seungyeop Choi (Yonsei University, South Korea)

Structural and Interfacial Engineering of Ceramic-Coated
Separators for Stable Lithium Metal Batteries

16:05 – 16:20 **General**

Jaejin Lim (Yonsei University, South Korea)

Decoding Electrode Microstructure-Performance
Relationships through FIB-SEM-Based 3D Model

16:20 – 16:35

Liang-Ting Wu (National Taiwan University of Science and Technology, Taiwan)

Unveiling Enhanced Li-Ion Mobility in Amorphous SEI Layers: A Machine-Learning-Accelerated Molecular Dynamics Study of the Solid Electrolyte/Li-Metal Interface

16:35 – 16:50 **General**

Yanyun Zhang (Ocean University of China, China)

Addressing Electro-Chemo-Mechanical Coupling Failure at the Sodium|Na₃PS₄ Interface towards Durable All-Solid-State Sodium Metal Batteries

Chaired by Shuai Yuan

16:50 – 17:05 **General**

Hongkai Hu (Ocean University of China, China)

Ionic Microenvironment Regulator Enables Enhanced Anion Trapping in Cationic Poly(ionic liquid) Solid Electrolytes for Low-Temperature Lithium Metal

17:05 – 17:20 **General**

Kiruba Muniyandi (University of Ulsan, South Korea)

Engineering Layered Zn(002) via Oscillation-Regulated Electrodeposition on Reconstructed Zn Substrates for Stable Aqueous Zinc-Ion Batteries

17:20 – 17:35 **General**

Closing

18:30 –

Dinner

Thursday 2 October 2026

Room: 1

Open discussion & networking

Poster presentation

Main Hall

Chaired by Prof. Seung-Taek Myung and Prof. Dr. Payam Kaghazchi

Monday 28 September 10:00 - Thursday 1 October 16:00

26-001

Tzu-Yu Kuo (National Yang Ming Chiao Tung University, Taiwan)

High-Energy-Density Solid-State Lithium Batteries Enabled by Ultrathin Composite Solid Electrolyte and High-Loading Composite Cathodes

26-002

Garam Lee (Korea Research Institute of Chemical Technology, South Korea)

Spray-Dried Microsized Porous Carbon Host with C-Doped g-C₃N₄ for High-Performance Sulfur Cathodes

26-003

Ramkumar Balasubramaniam (University of Ulsan, South Korea)

Theoretical and Experimental Investigation of W-Substituted P2-Type Na_{0.75}Mn_{0.60}Ni_{0.30}Fe_{0.10}O₂ Cathode Materials for Sodium-Ion Batteries

26-004

Jeong Jun Park (Korea Research Institute of Chemical Technology, South Korea)

MOF-Derived Co₂P-Embedded CNF Interlayers for High-Performance Lithium-Sulfur Batteries

26-005

Jun Ho Shin (Korea Research Institute of Chemical Technology, South Korea)

Multifunctional Roles of Oxide Additives in Dry Electrodes

26-006

Shaoning Zhang (Kyoto University, Japan)

Reliable Evaluation of Materials in Na-Ion Batteries Using NASICON Electrode

26-007

Ganeshbabu Mariappan (University of Ulsan, South Korea)

Correlating Structure and Na-Ion Diffusion in Ti/Mg Co-Doped P2-Type Na_{0.67}Mn_{0.60}Ni_{0.20}Fe_{0.20}O₂ Cathodes through Experimental and First-Principles Studies

26-008

Kyung woo Son (Korea Research Institute of Chemical Technology, South Korea)

Development of an elastic polymer protective layer incorporating crosslinker-functionalized inorganics for Li-metal batteries

26-009

Sanghyeock Jeong (Hoseo University, South Korea)

A Study on the Dry Fabrication Process of High Energy Density Anodes for LFP Batteries

26-010**Jae Kook YOON** (CPS Inc., South Korea)

Li-PAN/PAA Ionic-Conductive Binder Additive for Enhanced Cycling Stability and Rate Capability of Si/C Anodes and LFP Cathodes for LIB

26-011**Seung Hwa oh** (Korea Research Institute of Chemical Technology, South Korea)

Structural Stabilization of Silicon Anodes Using Spray-Drying Si/C/CNT Composite

26-012**Jinkyu Park** (Korea Research Institute of Chemical Technology, South Korea)

Suppressing PTFE Reduction through Electron-Blocking Ion-Permeable Layer in Dry Anodes

26-013**Junyoung Choi** (Korea Research Institute of Chemical Technology, South Korea)

Development of a Metal-Oxide/Organic Hybrid Protective Layer for Dendrite-Free Lithium metal anode

26-014**Junhee Lee** (Korea Research Institute of Chemical Technology, South Korea)

Enhanced Electrochemical Performance of Over-Lithiated Layered Oxide Cathodes through Co Incorporation while Maintaining the Li-to-Transition-Metal Ratio

26-015**Ho Jin Lee** (Korea Research Institute of Chemical Technology, South Korea)Microstructural Regulation and Enhanced Lithium-Ion Transport in Dry-Processed Thick NCM Cathodes via BaTiO₃ Addition**26-016****Junhee Lee** (Korea Research Institute of Chemical Technology, South Korea)Enhanced Rate Capability of LiFePO₄ Cathodes at Room and Low Temperatures through Ti Doping and Plasma Surface Modification**26-017****Ho Jin Lee** (Korea Research Institute of Chemical Technology, South Korea)Enhanced Interfacial Adhesion and Thermal Stability of Aluminum Current Collectors for Dry-Processed NCM Cathodes via a P3HT-CNT-BaTiO₃ Primer Layer**26-018****Jae Hyeon Jo** (KEPCO Research Institute, South Korea)

Hollandite-type Vanadium Oxyhydroxide with High Energy Density as a Cathode Material for Lithium-Ion Batteries

26-019**Jaesub Kwon** (Pohang University of Science and Technology (POSTECH), South Korea)

Understanding the Interfacial-Mechanical Degradation Feedback in High-Voltage Single-Crystal Mid-Ni Layered Oxide Cathodes

26-020

Jihee Choi (Korea Research Institute of Chemical Technology, South Korea)
Fluorinated Polymer Electrolytes with Enhanced Flame Retardancy and Wide-Temperature Electrochemical Performance

26-021

Ji Won Seo (Korea Research Institute of Chemical Technology, South Korea)
Synergistic Dual-Plasticizer Design for Balancing Ion Transport and Network Integrity in Gel Polymer Electrolytes

26-022

Seong-Jun Kwon (Korea Research Institute of Chemical Technology, South Korea)
Effect of Vacuum Sealing Conditions on Electrolyte Composition, Anode Interface, and Cycle Life in Lithium Metal Battery Pouch Cells

26-023

Jeong-Min Kim (Korea Electrotechnology Research Institute (KERI), South Korea)
Enhancing the Electrochemical and thermal stability of Ultrahigh-Nickel $\text{LiNi}_{0.96}\text{Co}_{0.02}\text{Mn}_{0.02}\text{O}_2$ Cathodes via Mechano-fusion-Assisted Li-Zr-O surface modification

26-024

Seong A Park (Korea Research Institute of Chemical Technology, South Korea)
An Al_2O_3 /PSSLi-Coated Separator for Enhanced Li^+ Transport and Long-Term Cycling Stability of Lithium Metal Batteries

26-025

Han Wool Jo (Korea Research Institute of Chemical Technology, South Korea)
Sublimation-Driven Pore Architecture in Dry-Processed Thick Cathodes for High-Rate, Long-Life Lithium-Ion Batteries

26-026

Han Wool Jo (Korea Research Institute of Chemical Technology, South Korea)
Reactive Sulfur-Embedded Carbon Additives for Enhanced Cycling Stability and Thermal Safety of Oxygen-Releasing Layered Cathodes

26-027

Jong Hyeok Yun (Korea Research Institute of Chemical Technology, South Korea)
Mitigating Mn Crossover and Interfacial Degradation in Sodium-Ion Batteries via a Multifunctional Ceramic-Coated Separator

26-028

Bitna Kima (Korea Electrotechnology Research Institute (KERI), South Korea)
Interfacial Chemo-Mechanical Degradation Behavior of Layered Oxide Cathodes in Sulfide- and Halide-Based All-Solid-State Batteries

26-029

Jemin Lee (Kyungpook National University, South Korea)

Dual-Anion Locally Concentrated Ionic Liquid Electrolytes for Enhanced Ion Transport and Stable Interphase Formation in Thin Lithium Metal Batteries

26-030

Jin Sik Nam (Korea Research Institute of Chemical Technology, South Korea)
A Sustainable One-Pot Spray-Drying Strategy for Fabricating High-Capacity LiFePO_4 Thick Dry Electrode via Nanoparticle Reconstruction

26-031

Eunbin Jang (Kyungpook National University, South Korea)
Electric-double-layer ordering couples dynamic anion replenishment with F-rich interphase formation in thin lithium metal batteries

26-032

Yoonkyo Seo (Ajou University, South Korea)
Nanochannel Orientation-Engineered Mesoporous Carbon Spheres for High-Power Potassium-Ion Hybrid Supercapacitors

26-033

Gyeongbeom Ryoo (Korea Electrotechnology Research Institute, South Korea)
Sequential Dual-Interfacial Engineering of High-Nickel Cathodes with Li_3PO_4 and SWCNTs for high-performance lithium-ion batteries

26-034

Junyoung Heo (Korea Electrotechnology Research Institute, South Korea)
Sulfur-Assisted Pore Reconstruction in Dry-Processed Cathodes for Practical Lithium–Sulfur Batteries

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Decoding and Controlling Interfacial Dynamics for Next-Generation Lithium–Sulfur Batteries

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Lithium–sulfur (Li–S) batteries offer compelling opportunities for high-energy-density and cost-effective energy storage, yet their practical implementation remains limited by complex and strongly coupled processes involving sluggish sulfur redox kinetics, polysulfide transport, interfacial degradation, and poor lithium reversibility. Addressing these challenges requires not only new materials but also a mechanistic understanding of how reactions and degradation evolve across the entire cell. In this talk, I will present our recent efforts to combine multimodal operando characterization with rational materials and interface design to understand and control these processes. First, the origin of the large activation barrier associated with Li_2S oxidation is investigated using operando transmission X-ray microscopy, confocal optical microscopy, electrochemical impedance spectroscopy, isothermal microcalorimetry, and synchrotron-based techniques. Our results reveal that sluggish Li_2S kinetics and the rapid dissipation of generated polysulfide intermediates from the cathode suppress the local chemical pathways required for efficient Li_2S conversion. Subsequent cycling transforms Li_2S into smaller and less crystalline structures with improved reaction kinetics, enabling greater local retention of polysulfides and eliminating the initial activation barrier. These mechanistic insights further suggest electrolyte strategies that regulate polysulfide generation and transport. These concepts are extended toward anode-free Li–S batteries, where zero excess lithium places stringent demands on lithium reversibility and current-collector stability. A self-healing GaIn-modified Cu current collector provides uniform lithium nucleation while mitigating polysulfide-induced Cu corrosion and dendritic lithium growth. Operando imaging, spectroscopy, and heat-evolution measurements reveal the underlying alloying/dealloying behavior and suppression of parasitic reactions, demonstrating how current-collector engineering can simultaneously address lithium deposition and corrosion. Together, these studies illustrate a pathway from observing dynamic battery chemistry to identifying hidden degradation mechanisms and ultimately designing more resilient interfaces.

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Low-Pressure, Low-Cost All-Solid-State Batteries Enabled by Monomer-in-Salt Electrolytes

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All-solid-state batteries (ASSBs) offer a promising route toward safer, high-energy-density energy storage, but their practical deployment is hindered by high stack-pressure requirements and costly manufacturing. Polymer electrolytes can alleviate these limitations through compliant interfacial contact and compatibility with scalable processing. However, conventional polyethylene oxide (PEO)-based electrolytes suffer from limited salt dissociation, crystallinity, segmental-motion-dependent Li^+ transport, and low Li^+ transference numbers.

This presentation introduces monomer-in-salt electrolytes as a “Beyond PEO” platform for low-pressure, low-cost ASSBs. In this inverted architecture, concentrated lithium salts form continuous ion-transport pathways, while a small fraction of polymerizable monomers provides mechanical stabilization through in situ polymerization. The resulting salt-rich polymer networks combine efficient Li^+ transport with conformal electrode contact, thereby reducing reliance on externally applied stack pressure.

Related systems, including conflicting-entropy-driven zwitterionic polymer electrolytes and ionic dimer elastomers, demonstrate how supramolecular organization and dynamic ionic bonding can reconcile ion conduction with mechanical resilience. Extending these principles to composite cathodes, monomer-in-salt catholytes are formed directly within dry-processed electrodes, creating solvent-free, compositionally uniform, and ionically percolated networks for Ah-class pouch cells.

By maintaining intimate interfaces under practically relevant low stack pressure, monomer-in-salt electrolytes reduce the need for pressure-intensive cell hardware. Their solvent-free processing, in situ catholyte formation, simplified electrode integration, and compatibility with existing lithium-ion battery manufacturing also provide a pathway toward lower production costs. These results establish monomer-in-salt electrolytes as a scalable platform for low-pressure, low-cost polymer-based ASSBs.

How to improve the performance of $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$: the point of view from crystal structure to electrolyte additive

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Spinel $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ (LNMO) represents a prominent next generation cathode active material for Li-ion batteries. However, severe parasitic electrode–electrolyte reactions at high voltage primarily remain a critical challenge to its commercialization. Single-crystal LNMO has been proposed as an effective strategy to mitigate this interfacial degradation. Here, we report a straightforward and cost-effective molten-salt synthesis for transforming polycrystalline LNMO into well-defined single crystals with highly exposed $\{111\}$ facets using alkaline molybdate (A_2MoO_4) fluxes. As a result, the optimized single-crystal morphology delivers superior cycling stability (34.3% at 500th cycle) and rate capability (48.3% at 10 C) over the polycrystalline LNMO with suppressed interfacial side reactions. In addition, LNMO also operates at high voltage ($\sim 5\text{V}$). Normally, there are $Fd\bar{3}m$ and $P4_332$ space group types of LNMOs. However, both LNMOs suffer phase segregation at intragranular side which affects the lithium diffusion pathway and structural degradation during cycles. On the other hand, Jahn-Teller distortion on Mn^{3+} as well as HF effects from electrolyte oxidation at high voltage window triggered lead Mn dissolution from LNMO that causes performance fade. In this work, a new salt is developed, lithium 1-fluoro benzimidazole (Li-1FB) for LNMO with two different space groups. Ex-situ XRD results show the Li-1FB postpones the formation of intermediate phase transition on both LNMOs at different states of charge. This new salt provides dramatic improvements, and it is suitable for further high-energy applications.

Impact of Composition and Morphology on the Electrochemical Performance of Li-Rich Layered Oxides

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Li-rich layered oxides are promising cobalt-free cathode materials for next-generation lithium-ion batteries because they can deliver high reversible capacities through combined transition-metal and oxygen redox reactions.¹ However, their electrochemical performance is strongly affected by Li excess, transition-metal composition,² and particle morphology, which influence reaction kinetics and structural stability during cycling.

In this study, Li-rich layered oxides, $\text{Li}[\text{Li}_x(\text{Ni}_{1-y}\text{Mn}_y)_{1-x}]\text{O}_2$ ($y = 0.50, 0.62$ and 0.75), were synthesized by solid-state reactions to investigate the effects of composition on the structure and electrochemical properties. Synchrotron X-ray diffraction confirmed the formation of Li-rich layered structures with characteristic Li/TM ordering. Electrochemical measurements revealed oxygen-redox activity associated with a voltage plateau near 4.55 V. Among the investigated compositions, samples with moderate Li excess exhibited the highest initial discharge capacity ($\sim 233 \text{ mAh g}^{-1}$), whereas excessive Li content led to capacity loss, indicating sluggish reaction kinetics and increased structural rearrangement.

To improve structural stability, particle morphology was controlled through a multi-step lithiation process using $\text{Li}_{1.2}\text{Ni}_{0.2}\text{Mn}_{0.6}\text{O}_2$ as a model composition. The resulting material exhibited enlarged primary particles and a reduced specific surface area compared with conventionally synthesized samples. Although the initial capacity was slightly lower, the morphology-engineered sample showed significantly improved capacity retention and suppressed voltage decay at 45 °C.

These results demonstrate that both composition and particle morphology play key roles in determining the capacity and stability of Li-rich layered oxides. While Li excess governs oxygen-redox utilization and reversible capacity, particle morphology strongly influences interfacial stability and degradation behavior. Understanding and controlling these factors is essential for the development of durable, high-energy-density lithium-ion battery cathodes.

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Formation Mechanism of Radially Oriented Microstructures in Tungsten-Doped Ni-Rich Layered Oxides Cathodes

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The microstructural evolution of Ni-rich layered oxide cathodes during calcination plays a decisive role in determining their structural stability and electrochemical durability. Nevertheless, the mechanisms by which high-valence dopants regulate crystallographic growth remain largely unexplored. Herein, we demonstrate that tungsten functions beyond a conventional lattice dopant by forming nanoscale Li_2WO_4 domains in $\text{LiNi}_{0.96}\text{Co}_{0.03}\text{Mn}_{0.01}\text{O}_2$ (NCM96) cathodes during high-temperature sintering. Atomic-resolution TEM revealed nanoscale Li_2WO_4 domains associated with specific crystallographic facets of NCM96, suggesting their role in directing facet-selective crystal growth during calcination. Such Li_2WO_4 nanodomains are proposed to modulate anisotropic crystal growth, thereby suppressing primary-particle coarsening and promoting the formation of a refined microstructure while preserving the layered framework. The resulting microstructure effectively mitigated the H2–H3 phase transition during cycling, thereby resulting in an initial discharge capacity of 234.3 mAh g⁻¹ at W-doped NCM96 and an improved capacity retention of 87.3% after 100 cycles, in comparison with 73.1% for the pristine NCM96 cathode. These findings establish Li_2WO_4 nanodomain-mediated facet engineering as an effective strategy for directing crystal-growth pathways and designing structurally robust Ni-rich layered cathodes.

Synergistic Modulation of Interfacial Chemistry and Residual Solvent Chemistry in Composite Solid Electrolytes

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Composite solid electrolytes (CSEs) integrating the flexibility of poly(vinylidene fluoride)-co-hexafluoropropylene (PVDF-HFP) with the mechanical strength of $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$ (LLZTO) are promising candidates for all-solid-state lithium metal batteries (ASSLBs).¹ However, their practical application is hindered by sluggish Li^+ transport kinetics caused by strong ion-solvent/polymer coordination and unstable electrode/electrolyte interfaces due to residual solvent decomposition. To overcome these bottlenecks, we present a dual-level strategy optimizing both the filler-polymer interface and the bulk solvation environment.

First, we engineered the surface of LLZTO fillers by constructing an amorphous $\text{Li}_x\text{TaOF}_{5-x}$ (LTOF) oxyhalide ion bridge via an in-situ reaction.² This LTOF layer acts as a competitive coordination site, weakening the Li^+ interaction with anions and polymer chains, thereby lowering salt dissociation energy. Simultaneously, it suppresses electron localization and prevents PVDF-HFP dehydrofluorination, enhancing interfacial compatibility. The resulting CSE achieves an ionic conductivity of 1.21 mS cm^{-1} and a Li^+ transference number of 0.58, enabling stable Li plating/stripping at 0.8 mA cm^{-2} for over 1100 hours.

Second, we addressed the fundamental paradox of residual solvents by introducing cyclopentyl methyl ether (CPME) to regulate the solvent chemistry. Through solvent-solvent dipole-dipole interactions, CPME modulates the electron cloud density of Dimethylformamide (DMF), systematically weakening its strong interactions with Li^+ , polymers, and fillers. This promotes the formation of anion-dominated solvation structures and a conjugated solvation network, significantly reducing the desolvation energy barrier. The optimized CSE (CSE-DCP) exhibits a high ionic conductivity of 1.63 mS cm^{-1} and a transference number of 0.81. Consequently, full cells with LiFePO_4 and $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ cathodes demonstrate exceptional cycle life (up to 4000 cycles at 5 C) and achieve a high energy density of 306 Wh kg^{-1} in pouch cells.

Together, these studies provide a comprehensive framework for designing high-performance CSEs by simultaneously engineering filler/substrate interfaces and bulk solvation environments, paving the way for practical, high-energy-density solid-state batteries.

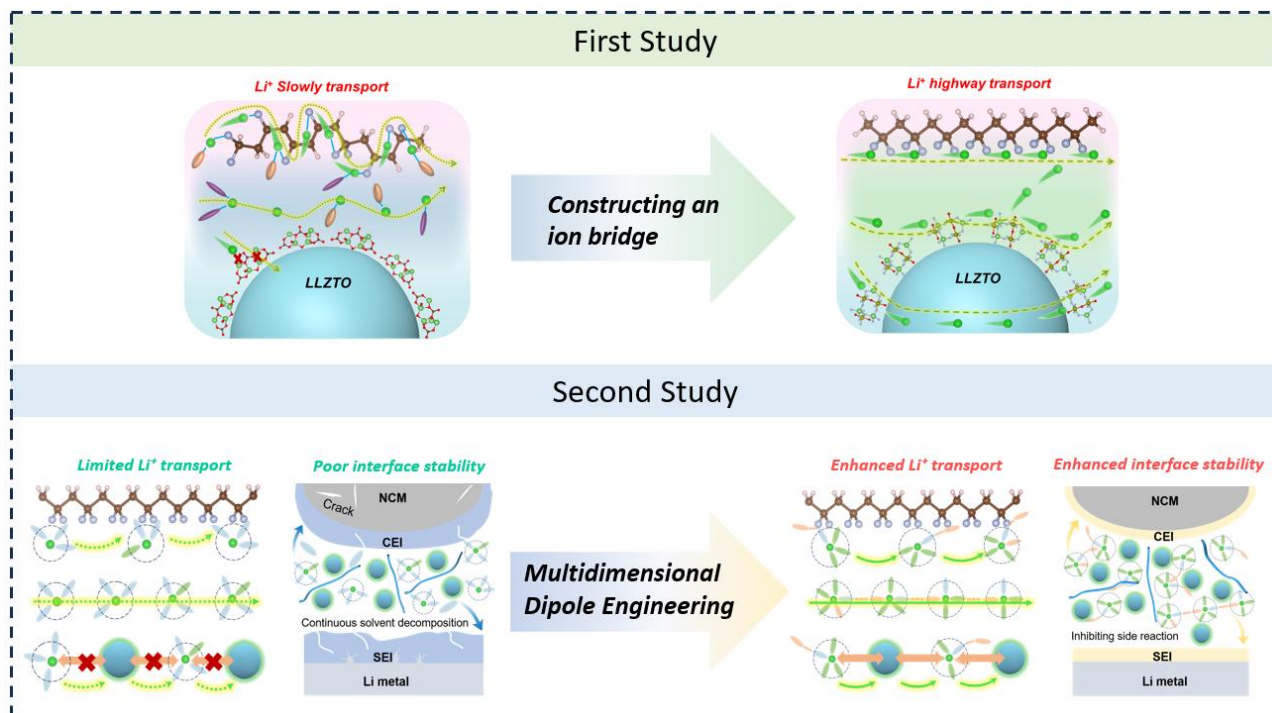


Fig 1. The schematic of modulation of interfacial chemistry and residual solvent chemistry.

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Visualizing and Unraveling Charge Transport in Battery Electrodes

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Lithium-ion and electronic transport within battery electrodes plays a decisive role in governing the rate capability and dynamic polarization of lithium batteries. In practical battery electrodes, charge-carrier fluxes operate under non-uniform driving forces across intricate multi-phase networks comprising electroactive NCM particles, conductive additives, and electrolytes. However, the intrinsic structural complexity, spatial tortuosity, and interfacial heterogeneities within these electrode systems obscure local potential drops, making it exceptionally difficult to resolve the physical origins of kinetic resistance during operation.

To resolve these transport bottlenecks, we present a combined experimental and theoretical framework that integrates 3D X-ray imaging with physical modeling. We first employ transmission X-ray microscopy (TXM) and projection X-ray microscopy (PXM) to reconstruct the three-dimensional microstructure of practical NCM composite electrodes, enabling direct visualization of operational concentration gradients and carrier propagation dictated by particle morphology, dimensions, and spatial connectivity. By coupling these quantitative structural parameters with an augmented transmission-line model and electrochemical impedance spectroscopy, we explicitly account for the interplay between geometric confinement and carrier-selective heterointerfaces. This integrated approach uncovers the physical origin of non-ideal impedance spectra, predicts rate-dependent lithium redistribution, and enables the direct spatial decoupling of intrinsic charge-transfer reaction kinetics from ion and electron transport overpotentials.

Controlling Lithiation Pathways for Low-Defect Ni-Rich Layered Cathodes

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Ni-rich layered oxides are leading candidates for high-energy lithium-ion batteries, yet their practical implementation remains constrained by structural degradation originating from defects generated during both synthesis and electrochemical cycling. In particular, deviations in lithium stoichiometry, heterogeneous lithium distribution, pore formation, transition-metal migration, and surface–bulk structural mismatch can strongly influence the electrochemical stability of these materials. Understanding how such defects develop during lithiation is therefore essential for establishing reliable strategies for the synthesis of structurally robust Ni-rich cathodes.

In this presentation, we discuss how precursor chemistry, lithium stoichiometry, particle architecture, and reaction atmosphere regulate lithiation pathways and defect evolution in Ni-rich layered oxides. Synchrotron X-ray diffraction and absorption spectroscopy, neutron diffraction, and complementary microstructural analyses are combined to correlate lithium distribution, transition-metal electronic structure, local coordination, pore evolution, and structural homogeneity with electrochemical performance. Case studies involving Li off-stoichiometry, particle-size and lithium-concentration gradients, and controlled pore evolution are presented, together with ongoing work examining how distinct lithiation pathways influence defect formation during LiNiO_2 synthesis. Collectively, these results establish lithiation-pathway control as a unifying strategy for minimizing atomic- and nanoscale defects and improving the cycling durability of Ni-rich layered cathodes.

Sulfonimides as Negative Charges for Advanced Battery Electrolytes

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Nonaqueous electrolytes are critical components for building high-performance rechargeable batteries. Lithium hexafluorophosphate (LiPF₆) has been the most prevailing conducting salt since the debut of lithium-ion batteries in commercial market in the 1990s; yet, the hexafluorophosphate anions tend to be decomposed over long-term battery operations, calling for the design of new anions with enhanced chemical and electrochemical performances [1].

Sulfonimide anions [$-\text{SO}_2-\text{N}^{(-)}-\text{SO}_2-$] are featured with two sulfonyl groups in conjugation with imide linkage, in which the negative charges are highly delocalized over one nitrogen and four oxygen atoms. Sulfonimide-based alkali metal salts and ionic liquids generally show better chemical stability against the classic hexafluorophosphate-based ones, thus becoming tantalizing candidates for liquid, gel/plasticized, and composite polymer electrolytes [2].

In this talk, we will present recent advances related to structural design and property-regulations of sulfonimide anions, with due attention paid to the unique impact of super-delocalized anions and their synergy with inorganic fillers in composite polymer electrolytes [3,4]. We will also discuss possible strategies on tailoring sulfonimide anions for future solid-state batteries.

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Oxygen vacancies engineering to boost Li-ion transport and cycling stability in TiNb_2O_7 anodes

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With the rapid growth of portable electronics and electric vehicles, lithium-ion batteries (LIBs) have become a core technology for modern energy storage systems. However, the increasing demand for lithium resources, coupled with rising extraction costs, poses significant challenges to the sustainable development of LIBs. TiNb_2O_7 (TNO) has attracted attention due to its intercalation-type mechanism, moderate operating voltage and a theoretical capacity of $\sim 387 \text{ mAh g}^{-1}$. This study investigates the effects of Sc^{3+} doping on enhancing the structural and electrochemical properties of TNO (Fig. 1). A combination of analytical techniques, including DFT calculations and pseudocapacitive analysis, is employed to elucidate lithium-ion storage behavior. 3 mol.% Sc^{3+} doping achieved the highest reversible capacity of 363.9 mAh g^{-1} at 0.1C, with a capacity retention of 86.6% after 500 cycles at 5 C. CV, GITT, and EIS tests all showed improved lithium-ion diffusion kinetics and charge transfer properties at this doping level. This study demonstrates that targeted compositional modifications can significantly optimize the structural features and electrochemical behavior of TNO, providing a promising pathway for developing high-rate, long-cycle lithium-ion battery anode materials.

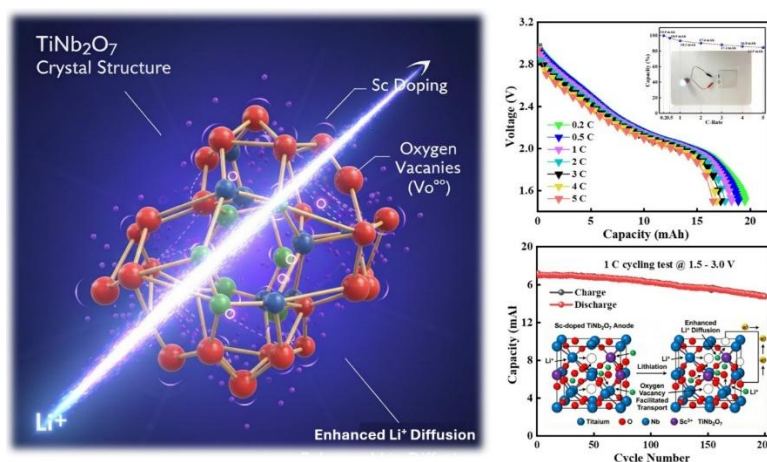


Fig. 1 Discharge curves of pouch cell by using NCM/TNO:Sc full cell; Inset: LED lighting; Cycle life tests of NCM/TNO:Sc full cell. Inset: The corresponding theoretical calculations.

Halide-based Electrolytes for All Solid-State Batteries

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All-state-state lithium batteries (ASSLBs) have gained worldwide attention because of the high ionic conductivity of SEs, intrinsic safety and increased energy density. In ASSLBs, solid-state electrolyte is a key component and interface is a big challenge. In this talk, I will report recent progresses of halide-based electrolytes and electrodes.

Compared with sulfide-based electrolyte, both experimental and theoretical results recently demonstrate that halide-based electrolytes have more advantages including high RT ionic conductivity ($>10^{-2} \text{ S cm}^{-1}$), wide electrochemically stable window, high air-stability, good stability toward oxide cathode materials, and even salable water synthesis strategy. Furthermore, we will discuss electrode fabrication. In the end, the perspectives will be given.

Precision Assembled Single Layer Pouch Cells with Reference Electrode for Studying Emerging Battery Chemistries

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Precise and reproducible electrochemical testing of lithium-ion battery materials is essential for both academic research and industrial development. While coin cells are widely used due to their simplicity and accessible assembly method, they offer limited control over critical variables such as electrode alignment, pressure distribution, electrolyte distribution and reference electrode placement. These factors can introduce variability into electrochemical measurements and make it difficult to distinguish the behaviour of individual electrodes within a full cell.

To address these limitations, we present the continued development of a precision assembled, single layer pouch (SLP) cell architecture incorporating various types of lithium reference electrode for three electrode electrochemical testing. The work was enabled through the use of a precision automated cell assembly system, allowing electrode, separator and reference electrode placement to be controlled reproducibly while reducing operator dependent variation. A circular pouch cell format was used to retain the convenience and material efficiency of conventional coin cell testing while providing greater control over cell geometry, stack pressure and reference electrode integration.

This work focuses on the development and validation of lithium reference electrode configurations within NMC/graphite full cells. Different separator materials, separator geometries, reference electrode positions and electrolyte distributions were investigated to identify configurations capable of providing stable and representative measurements of individual electrode potentials. The resulting three electrode architecture enables simultaneous measurement of full cell voltage together with the independent cathode and anode potentials during formation and subsequent cycling.

Reference electrode stability was evaluated over extended cycling and at different states of charge. The developed configuration demonstrated stable electrode resolved voltage measurements, with reference drift as low as 0.76 mV over a period of five days at the upper state of charge. Greater drift was observed at lower states of charge, demonstrating the importance of validating reference electrode behaviour across the complete operating voltage range rather than assuming constant stability throughout cycling.

The electrochemical behaviour of the three electrode cells was compared with both equivalent two electrode precision assembled cells and conventional coin cells. This comparison was used to assess whether incorporation of the reference electrode significantly perturbs normal cell behaviour, while

also examining differences associated with cell format and assembly method. Comparison with coin cells provides an important benchmark for translating established laboratory testing methods into a more controlled three electrode architecture.

The developed cells were subsequently used for electrochemical impedance spectroscopy, enabling the impedance response of the cathode and anode to be resolved independently before cycling, during formation and following subsequent cycling. This provides additional insight into the evolution of electrode specific resistance and degradation mechanisms that cannot be obtained from conventional two electrode full cell measurements alone.

This work demonstrates a practical approach to reproducible three electrode battery testing in a format suitable for routine laboratory use. By combining controlled automated assembly, defined stack pressure and validated lithium reference electrode placement, the method provides a bridge between conventional coin cell testing and more advanced electrode resolved electrochemical analysis. The approach may support more reliable interpretation of full cell behaviour and enable deeper investigation of degradation mechanisms in emerging battery materials and chemistries.

Asymmetric Li Insertion/Extraction

Kinetics of $\text{Li}[\text{Li}_{1/3}\text{Ti}_{5/3}]\text{O}_4$

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The reaction kinetics of lithium insertion materials have been investigated in our laboratory using the dilute-electrode method [1-4], which enables the intrinsic reaction kinetics of Li insertion materials to be evaluated with minimal influence from ionic and electronic transport within porous composite electrodes. In this study, kinetic asymmetry between Li insertion and extraction in $\text{Li}[\text{Li}_{1/3}\text{Ti}_{5/3}]\text{O}_4$ (LTO) was investigated.

Figure 1 shows the kinetics of Li insertion and extraction measured by constant-voltage charge and discharge tests using a dilute LTO electrode containing 5 wt% active material. To quantitatively compare the reaction kinetics, the time required to reach half the reversible capacity, $t_{1/2}$ was evaluated. At low overvoltages, similar $t_{1/2}$ values were obtained for Li insertion and extraction. At high overvoltages, however, the $t_{1/2}$ values for Li extraction were much smaller than those for Li insertion, demonstrating that Li extraction in LTO proceeds much faster than Li insertion.

The dependence of $t_{1/2}$ on the applied voltage also differed markedly between the reactions. For Li insertion, $t_{1/2}$ remained nearly constant irrespective of applied voltage. In contrast, $t_{1/2}$ for Li extraction decreased with increasing overvoltage. These results suggest that kinetic asymmetry of LTO originates from differences in the rate determining processes or reaction mechanisms of Li insertion and extraction.

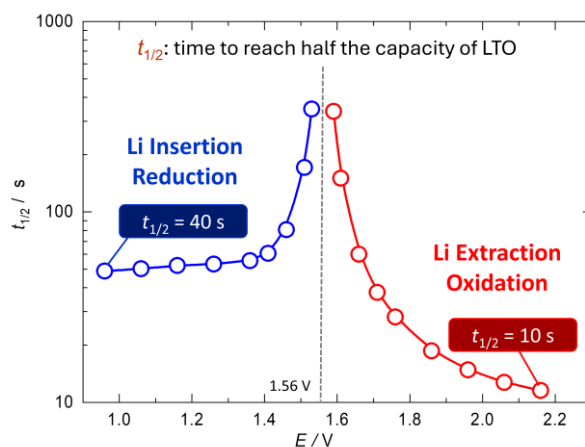


Fig. 1 time to reach half the capacity, $t_{1/2}$ as a function of applied potential for the dilute LTO electrode containing of 5 wt% active material.

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Direct Regeneration of Olivine Cathodes: LFP Successes and Emerging Challenges with LMFP and Blends

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The rapid deployment of Li-ion batteries creates a growing need for recycling strategies capable of preserving the functional value embedded in electrode materials. This is particularly relevant for olivine cathodes such as LiFePO_4 (LFP) and $\text{LiMn}_x\text{Fe}_{1-x}\text{PO}_4$ (LMFP), whose relatively low raw-material value can limit the economic relevance of conventional pyro- and hydrometallurgical approaches. Our work explores direct regeneration as a strategy based on identifying the dominant degradation mechanisms and selectively repairing spent cathode materials under mild conditions.

LFP represents a particularly favorable case. Significant capacity loss may occur while the olivine framework remains preserved, with lithium inventory loss being a major degradation mechanism. Based on this observation, several room-temperature relithiation strategies were developed, progressing from LiI-based chemical lithiation in organic media to a rapid solvent-free process and, more recently, to an aqueous route combining a lithium source with ascorbic acid. This approach was further validated on severely swollen 50 Ah commercial LFP cells. Despite electrolyte degradation, gas generation, dry-out and heterogeneous lithium loss, the LFP structure remained largely preserved and could be restored by a mild aqueous treatment at the 100 g scale and validated by the production of 18650 cells containing 100% regenerated active material.

Extending this concept to LMFP reveals additional limitations. Relithiation remains highly effective when lithium loss mainly involves Fe oxidation, whereas deeper delithiation involving Mn oxidation leads to incomplete regeneration due to manganese dissolution and irreversible local modifications. Finally, post-mortem analysis of commercial LMFP/NMC blended cells highlights further complexity, including transition-metal dissolution, cathode–anode cross-talk and structural degradation. Direct relithiation can largely restore the LMFP fraction, whereas the degraded NMC component cannot be fully repaired. These results show that direct recycling should be viewed, today, as a chemistry-specific diagnosis-and-repair strategy rather than a universal relithiation process.

Reinventing Electrolytes: Deep Eutectic Solvents for Energy Storage

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Abstract. Deep eutectic solvents (DESs) are emerging as versatile and sustainable electrolytes for electrochemical energy storage technologies. Their intrinsic tunability and favorable physicochemical properties make them attractive candidates for post-lithium batteries and supercapacitors. In this contribution, we correlate the structure–property relationships of thoughtfully designed DESs with their application in specific devices. Building on these findings, we discuss how DES composition influences conductivity, viscosity, redox stability, and device performance. More specifically, we developed a NaCl/glycerol-based eutectic capable of cycling in both supercapacitors and hybrid batteries, with electrochemical performance comparable to that of conventional reference electrolytes. To further expand the applicability of our mixtures, we replaced NaCl with $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ to obtain Zn-based, low-temperature transition mixtures for use in low-rate Zn-ion batteries. Overall, our work positions DESs as promising electrolytes for advanced energy storage systems and lays the groundwork for future research aimed at optimizing their structure–property relationships and integrating them into high-performance devices.

Acknowledgments: This work was supported by the Fondo Italiano per la Scienza (FIS) under the FIS3 programme, funded by the Ministry of University and Research (Ministero dell'Università e della Ricerca) - Project CUP: [B53C25003870001]. This study was carried out within the GENESIS project funded by the Ministero dell'Università e della Ricerca within the PRIN 2022 program.

AI-Accelerated Modelling and Design of Liquid Electrolyte for Rechargeable Batteries

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Electrolytes are central to rechargeable battery performance because they govern ion transport, solvation structure, electrochemical stability, and electrode-electrolyte interfacial reactions. However, electrolyte discovery remains challenging because the relevant phenomena span molecular chemistry, liquid structure, transport, polarization, and interphase formation. In this talk, I will discuss our recent work on AI-accelerated liquid electrolyte research along two connected directions: materials modelling and materials design.

For materials modelling, we developed PiNN, an equivariant neural-network suite for electrochemical systems [1], with modules for potential energy surfaces, molecular dipole and quadrupole moments [2], and charge-response kernels. Together with adaptive learn-on-the-fly workflows [3] and finite-field coupling [4], this enables machine-learning molecular dynamics simulations of liquid electrolyte in reactive and polarizable environments.

For materials design, we introduced a Generative Solvent Design System (GSDS) for rechargeable batteries [5], coupling a graph-based molecular generator with physics-informed machine-learning property predictors for redox stability, viscosity, melting point, donor number, and dielectric constant. This workflow moves beyond screening known molecules toward inverse design of electrolyte solvents, producing candidate fluorinated diluents and nonfluorinated weakly solvating solvents for alkali metal batteries.

Together, these studies show how AI can accelerate both the modelling and design of liquid electrolytes, while also highlighting the need for systematic property measurements and experimental validation data to make the best use of computational predictions.

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Assessment of Thin Lithium Metal Electrodes via Voltage Profile Diagnostics

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Li metal offers exceptionally high capacity and a low electrochemical potential, making it attractive for high-energy batteries [1]. Anode-free Li-metal batteries theoretically maximize energy density by eliminating preloaded Li, but their practical advantage is highly sensitive to irreversible Li loss and engineering penalties. Our previous studies showed that Thin-Li batteries can buffer such losses with only a small mass and volume penalty, thereby improving retained energy density. We further proposed capacity-matched Thin-Li as a benchmark for current-collector modifications, which must justify their added mass, thickness, electrolyte demand, and Li consumption [2]. Anode-free and Thin-Li batteries can therefore be regarded as complementary limited-Li configurations requiring highly reversible Li-metal cycling.

Assessment of preloaded Li-metal electrodes remains challenging. Conventional Cu|Li cells conveniently measure Coulombic efficiency but mainly reflect Li deposition on Cu rather than on a Li-metal surface. Cells containing a precisely limited Li reservoir better represent Li-on-Li cycling, but require difficult control of ultrathin Li loading and lengthy cycling until depletion. We therefore propose a rapid screening method based on voltage profiles from readily assembled Li|Li symmetric cells. A single plating–stripping cycle provides practical information on deposition quality and reversibility, enabling efficient comparison of Li electrolytes without precise control of the preloaded Li amount.

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Critical Role of the Current Collector for Lithium-Metal Electrodes

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Lithium-metal batteries are considered the next big step towards higher energy densities – especially when reducing the excess of lithium in the system to essentially zero. The greatest challenge in this regard is presumably the potential risk of dendritic lithium deposition and the high reactivity of lithium metal with all commonly known electrolyte systems, resulting in a continuous loss of electrochemically active lithium. A frequently overlooked source of such losses, however, is the interface with the current collector. In fact, classically used copper foils are considered “non-reactive” or “inert” in contact with lithium metal.

Herein, we will show that this is not the case, in fact, and that the interface with the current collector adds to the continuous loss of electrochemically lithium. Moreover, we present potential alternatives to the classically used copper foil, targeting a more favorable lithium stripping and plating behavior, while ideally also enabling substantially higher energy densities at the full-cell level.

Pilot-Scale Prototyping and Validation of Lithium-Ion Pouch Cells for Electric Vehicle Applications

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Bridging the gap between laboratory-scale battery research and industrial-scale manufacturing remains a major challenge for the commercialization of lithium-ion technologies. In this study, a pilot-scale lithium-ion battery production line is established and validated through the fabrication of large-format pouch cells and their integration into a functional electric vehicle (EV) battery system. The fully integrated manufacturing line includes slurry preparation, electrode coating, calendaring, cutting, stacking, electrolyte filling, sealing, and pouch cell assembly.

Lithium iron phosphate (LiFePO₄) cathodes and graphite anodes are optimized for scalable processing, with emphasis on electrode uniformity, mechanical integrity, and manufacturing reproducibility under high areal loading conditions. Using optimized materials and processing parameters, pouch cells with a nominal voltage of 3.2 V and a capacity of approximately 6 Ah are fabricated. The cells achieve a gravimetric energy density of approximately 130 Wh kg⁻¹ while exhibiting excellent batch-to-batch consistency and stable electrochemical performance. After 100 charge–discharge cycles, the cells retain approximately 97% of their initial capacity, demonstrating the robustness of both the electrode design and manufacturing process.

For practical validation, multiple pouch cells were assembled into a 12.8 V, 40 Ah prototype battery pack with dimensions comparable to a conventional automotive battery. The pack was installed in an electric vehicle platform provided by a car manufacturer in Kazakhstan and was successfully used as a direct replacement of a standard auxiliary battery to power onboard electronics and supports vehicle startup under real driving conditions.

These results demonstrate a complete pathway from pilot-scale manufacturing to real-world application, highlighting the importance of pilot production infrastructure in accelerating technology transfer and commercialization of advanced lithium-ion batteries for electric mobility.



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Current Collectors and Interfacial Engineering for Anode-Free Lithium Metal Batteries

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Anode-free lithium metal batteries offer high energy density but suffer from nonuniform lithium deposition, unstable interfaces, and rapid loss of active lithium. In this study, lithiophilic current collectors and interfacial engineering strategies were developed to regulate lithium nucleation and improve cycling stability. Copper substrates modified with $\text{Cu}_{1.94}\text{S}$ -based interfacial layers provided abundant nucleation sites and reduced the lithium nucleation overpotential from approximately 243 to 121 mV. These modifications promoted uniform lithium deposition, reduced polarization, and improved interfacial stability during repeated plating and stripping. When paired with an NCM811 cathode, the engineered current collectors delivered improved capacity retention compared with bare copper. These results demonstrate that controlling current collector surface chemistry is an effective strategy for enhancing lithium utilization and extending the lifetime of anode-free lithium metal batteries.

From Native Lithium Surface Chemistry to Temperature-Dependent SEI Evolution in Lithium-Metal Batteries

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Lithium-metal batteries (LMBs) are among the most promising next-generation energy storage technologies owing to their exceptionally high theoretical energy density. Despite major advances in electrolyte design, interfacial engineering, and cell architecture, the intrinsic instability of lithium metal remains a critical barrier to practical implementation. Research efforts have traditionally focused on the formation and evolution of the solid-electrolyte interphase (SEI) during electrochemical cycling. In contrast, the chemical and electronic state of lithium prior to cell assembly and operation has received comparatively limited attention.

This work examines how the native surface chemistry of lithium metal evolves upon exposure to technologically relevant gas environments and how these early-stage reactions influence subsequent interphase formation. Using ambient-pressure photoelectron spectroscopy together with complementary surface-sensitive techniques, the interactions of lithium metal with oxygen, carbon dioxide, and nitrogen are investigated, revealing distinct reaction pathways, surface passivation mechanisms, and modifications of the electronic structure. Emphasis is placed on the evolution of the lithium work function as a quantitative descriptor of the surface state and an indicator of the thermodynamic driving forces governing interfacial reactions.

Building on these insights, a broader framework is proposed in which lithium processing conditions define the native chemical and electronic state of the metal surface, quantified through its work function, which subsequently influences SEI nucleation and temperature-dependent interfacial evolution. Within this perspective, the work function emerges as a bridge between lithium surface chemistry and battery performance, offering new opportunities for predictive interface engineering in lithium-metal batteries. Attention is given to the influence of temperature on interphase composition, ion transport, and lithium deposition behaviour across a broad operating window.

These results suggest that control of the native chemical and electronic state of lithium may be as important as electrolyte design itself, providing a new paradigm for understanding and engineering lithium-metal interfaces across a wide range of operating temperatures.

Interfacial Engineering of Lithium Metal for High-Energy Batteries: Insights from Redox-Mediator-Based Li–O₂ Systems

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Lithium metal is a key anode candidate for next-generation high-energy-density batteries, yet its practical application is severely limited by interfacial instability, parasitic reactions, and uncontrolled reactions with soluble redox-active species. These challenges become particularly pronounced in Li–O₂ batteries employing soluble redox mediators, where mediator crossover can induce redox shuttling, active-material loss, and corrosion of the lithium-metal anode. Here, we present a series of material and interfacial strategies for regulating these dynamic processes and stabilizing lithium metal.

Functional separator modification using Mn_xCo_{3–x}O₄ promotes reversible iodide redox coupling while suppressing mediator crossover. Molecularly designed mediators, including benzyltriethylammonium iodide and 4-fluoro-2-iodoaniline, further couple Li₂O₂ oxidation with in situ formation of protective interphases. Complementary approaches based on AgTFSI pretreatment and polymeric ionic-liquid additives generate lithiophilic or ion-regulating surface layers that mitigate parasitic reactions and substantially enhance cycling stability. Operando synchrotron X-ray diffraction and X-ray computed tomography are employed to correlate mediator chemistry with lithium-interface evolution and cell degradation.

Together, these studies demonstrate that long-term stabilization of lithium metal requires coordinated regulation of redox-active species transport, separator functionality, and dynamic interphase chemistry. The insights obtained from Li–O₂ batteries provide broader design principles for controlling reactive lithium-metal interfaces in high-energy rechargeable battery systems.

Microstructure and electrochemical performance of Li-Na-Mg alloy anode for Li metal batteries

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Lithium metal and its alloys are attractive anode materials for next-generation rechargeable batteries owing to their high theoretical capacity and energy density. However, severe volumetric changes during cycling can compromise both durability and safety. Herein, porous Li–Na alloy and Li–Na–Mg alloy anodes are developed by selective lithium extraction to introduce internal free volume that buffers volume fluctuation and improves structural integrity. Full cells with the NMC622 cathodes demonstrate superior cycling stability compared with pure lithium anodes under low charging rates (0.1C), with the optimized cell retaining approximately 80% of its initial capacity after 120 cycles. In addition, coating the Li–Na–Mg alloy onto Cu foil reduces electrode thickness, enabling higher energy density and emphasizing the intrinsic role of the porous structure. Overall, the engineered porous architecture effectively enhances electrochemical performance and cycling stability, providing a promising strategy for high-energy-density lithium metal batteries.

Charge–Discharge Behavior of Li–Mg Alloy Electrodes in Ionic Liquid Electrolytes

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Lithium–magnesium (Li–Mg) alloys have emerged as promising lithium metal anodes owing to their enhanced cycling stability compared with pure lithium metal. Although improved cyclability has been reported for various battery systems, most previous studies focused on thick alloy electrodes containing excess lithium, leaving the performance of practically relevant thin foils insufficiently clarified.¹⁻²

In this study, Li–Mg alloy foils with a thickness of 20 μm were evaluated as negative electrodes in LiCoO_2 full cells employing ionic liquid electrolytes. The influence of Mg content on charge–discharge behavior and cycle life was systematically investigated.

The electrochemical performance strongly depended on Mg concentration. While pure lithium metal showed gradual capacity fading during cycling, Li–Mg alloy electrodes containing ≥ 3 wt% Mg exhibited significantly improved capacity retention. In cells with an areal capacity of 2 mAh cm^{-2} , the cycle number at 90% capacity retention increased from 164 cycles for lithium metal to 328 cycles for a Li–Mg(7 wt%) alloy electrode. These results demonstrate that appropriate Mg incorporation effectively stabilizes lithium deposition/dissolution processes and mitigates inactive lithium accumulation, leading to prolonged cycle life even in thin-foil configurations.

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Operando NMR spectroscopy for battery materials

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In recent years, the development of advanced rechargeable batteries, ranging from conventional lithium-ion batteries to emerging all-solid-state batteries, has increasingly relied on a detailed understanding of the reaction mechanisms of electrode and electrolyte materials.

Nuclear Magnetic Resonance (NMR) spectroscopy has become an powerful analytical tool in battery research because it enables the direct observation of lithium and sodium ions. Through the element-selective detection of these ions, NMR provides unique information on local structures, chemical states, and ion dynamics in battery materials. NMR has been successfully applied to a wide range of battery materials, including cathodes,[1,2] anodes,[3] and solid electrolytes,[4] providing insights that are often difficult to obtain by diffraction- or microscopy-based techniques.

As battery reactions continuously evolve during charge and discharge, operando characterization techniques capable of monitoring electrochemical processes under realistic operating conditions have become increasingly important. In parallel with advances in X-ray diffraction, electron microscopy, and other analytical methods, significant efforts have been devoted to extending NMR measurements from conventional ex-situ experiments to in-situ and operando investigations.

Historically, in-situ and operando NMR studies have relied on custom-designed probes and specialized electrochemical cells. Such approaches have enabled the observation of lithium intercalation/deintercalation in graphite[5] and silicon anodes[6], lithium metal deposition[7], dendrite formation, and electrochemical processes in all-solid-state batteries.[8] Despite their scientific value, however, these methodologies have remained largely limited to specialized research groups because of the substantial instrumentation and technical expertise required.

To facilitate the broader adoption of operando NMR in battery research, we recently introduced the first operando NMR probe for battery analysis developed by an NMR instrument manufacturer. In addition, in collaboration with Prof. Gotoh at the Japan Advanced Institute of Science and Technology (JAIST), we developed a novel electrochemical cell for operando NMR studies of all-solid-state batteries.

In this presentation, NMR spectroscopy for battery-material characterization and recent advances in in-situ and operando NMR methodologies will be introduced. Representative examples of operando NMR measurements, including lithium intercalation/deintercalation in graphite anodes (Fig.1) and lithium dendrite characterization, will be presented to illustrate the capabilities of the technique. The potential of recently developed operando NMR approaches for all-solid-state battery studies will also be discussed.

Phase-Field Modelling of Microstructural Evolution and Degradation in Battery Materials

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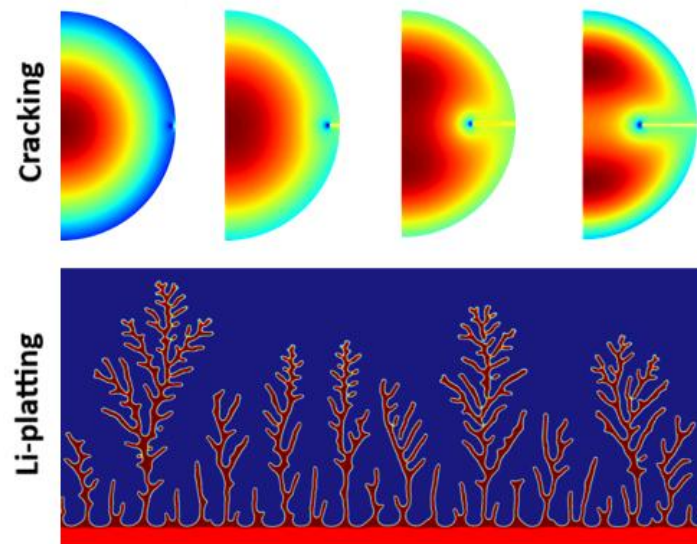
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Battery degradation is strongly influenced by local heterogeneities in transport, reaction kinetics, lithiation, and mechanical stress. In the mesoscale, microstructure-resolved models capture these effects by explicitly representing the solid, electrolyte, and interfacial domains of battery electrodes, revealing local current-density hot spots, evolving tortuosity, and non-uniform stress fields that are averaged out in conventional homogenized models. [1] However, these approaches become particularly challenging when degradation itself substantially changes the electrode microstructure, as occurs during crack propagation or lithium-metal deposition.

Phase-field methods provide a natural framework for addressing this limitation. By representing cracks, interfaces, and material phases through continuous field variables, they allow complex morphological evolution without requiring the evolving interfaces to conform explicitly to the computational mesh. [2, 3] In this talk, we present phase-field approaches for two highly relevant scenarios where microstructure changes play a role in battery degradation: (i) cracking of graphite particles driven by mechanical fatigue due to repeated cycling, and (ii) nucleation and growth of lithium-metal dendrites upon plating.

By coupling microstructural evolution with electrochemistry, transport, and mechanics, these models provide a route towards identifying the conditions that trigger accelerated degradation and towards more predictive descriptions of battery lifetime and safety.



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Developing a Cloud-Connected Platform for Energy Materials Acceleration

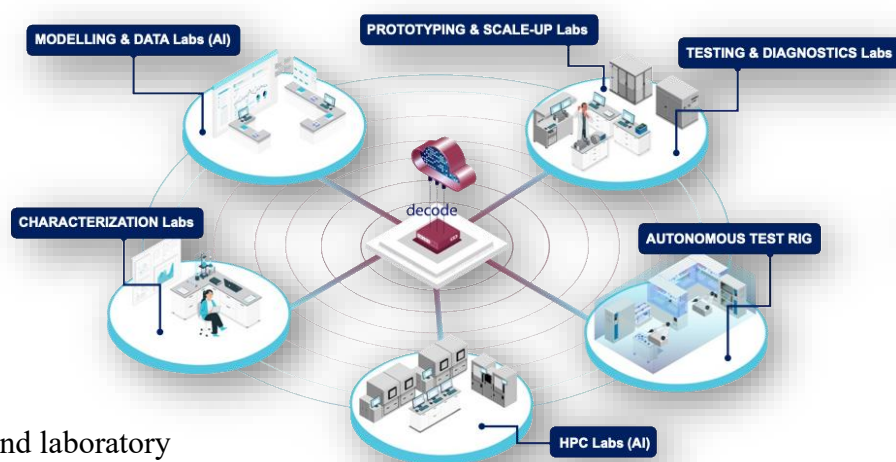
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Abstract. Research and development in the sustainable energy technology field is entering a transformative phase. In the future, progress in this field will be driven by advances in artificial intelligence, cloud computing, and laboratory



automation. This presentation reports on the advances made for developing a platform for cloud-connected labs. The decentralized, cloud-connected platform consists of three pillars: (i) collecting and standardizing methods and data, (ii) connecting laboratories and knowledge domains through semantic interoperability, and (iii) orchestrating multi-scale and multimodal research workflows using AI-powered digital agents. This framework provides a pathway to quantify efficiency gains in the R&D process itself and steer process orchestration towards optimal effectiveness and correspondingly reduced effort. This ambitious undertaking responds to the pressing need to accelerate the global energy transformation through strategies for the rapid development and scale-up of materials, while addressing technological sovereignty and supply chain security of key raw materials for a green economy. The imperative towards materials circularity adds further complexity, reinforcing the need to consider the entire value chain when designing new materials.

Synthesis and Characterization Technologies for Li₂S as a Key Raw Material in Sulfide-type Solid-State Electrolytes

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Sulfide solid-state electrolytes (SSEs) are widely regarded as the linchpin for the commercialization of all-solid-state batteries (ASSBs). However, the widespread application of sulfide SSEs is fundamentally bottlenecked by the high cost, safety concerns, and complex synthesis of their critical precursor, lithium sulfide (Li₂S). In this study, we systematically review the existing synthesis strategies for Li₂S and critically evaluate the most promising pathways toward low-cost, safe, and scalable mass production from an industrialization perspective, outlining their respective advantages and limitations. Building upon these evaluations, we introduce a novel solvent-free metathesis method for Li₂S synthesis. As recently demonstrated in our work (*Nat. Commun.* 2025, 16, 9981), this innovative approach exhibits exceptional potential for green, cost-effective, and large-scale manufacturing. Ultimately, this methodology provides a highly viable pathway toward the economically feasible production of sulfide SSEs, thereby accelerating the practical realization of high-performance ASSBs.

AI-aided Development of Low-Cost Halide Solid Electrolytes

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Halide solid electrolytes (HSEs) are promising for high-voltage cathodes, yet a combination of high ionic conductivity, robust Li metal compatibility, and cost-effectiveness, is fundamentally challenging to achieve. Here, we introduce a chemistry-driven paradigm, guided by machine learning, to unlock isotropic, three-dimensional (3D) superionic conduction and improve Li metal compatibility in HSEs. We identify a Ti/F co-doping strategy in the cost-effective Li_2ZrCl_6 (LZC) system, where substituting Zr with the more electronegative Ti induces a powerful inductive effect. This effect weakens Li^+ -anion coupling, thereby significantly reducing the *a-b* plane migration barrier and transforming the native 1D ionic transport into a 3D percolating network. The resulting electrolyte, $\text{Li}_{2.1}\text{Zr}_{0.9}\text{Ti}_{0.1}\text{Cl}_{5.7}\text{F}_{0.3}$, achieves a room-temperature ionic conductivity of 2.9 mS cm^{-1} . Additionally, it can generate a gradient interfacial passivation layer to stabilize the Li metal electrode for long-term cycling stability exceeding 2000 h at 1 mA cm^{-2} in Li symmetric cells. This unique combination of properties enables the all-solid-state Li metal batteries to realize stable cycling performance (86.5% capacity retention after 1000 cycles at 1 C in mold-type cells) and high areal capacity (3.6 mAh cm^{-2} in pouch-type cells), demonstrating a viable path towards practical, high-energy-density devices. This work establishes a new paradigm for rapidly and efficiently screening key target materials for practical device applications. An extension of this work identifies a P, S co-doped LZC, which retains similar cost to LZC but with a higher room-temperature ionic conductivity of over 1 mS cm^{-1} .

Universal Insights into Ion Transport in Solid-State Electrolytes

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The development of next-generation green-energy technologies—such as all-solid-state batteries, fuel cells, and electrochemical devices—critically hinges on our ability to design solid-state electrolytes (SSEs) with exceptional ionic conductivity. Yet, the design of high-performance SSEs remains a grand challenge due to the inherent complexity of these materials and the limited understanding of their atomistic transport mechanisms. In this talk, I will present a comprehensive, data-driven exploration of ion transport in inorganic SSEs, fusing insights from recent studies of ours that leverage advanced first-principles simulations, machine learning, and unsupervised analysis techniques [1,2].

First, I will introduce a universal perspective on SSE performance, built upon a curated database of zero- and finite-temperature quantum simulations covering over 60 diverse materials [1]. Through statistical correlation, principal component analysis, and clustering, we reveal that vibrational and elastic properties—especially those incorporating anharmonicity—are the most informative descriptors of SSEs, surpassing more conventional ones like chemical composition or defect energetics. These findings challenge prevailing assumptions and point toward a new classification framework based on lattice dynamics rather than ionic species.

Second, I will delve into the collective nature of ionic diffusion by presenting a novel unsupervised k-means clustering algorithm that identifies concerted hopping events from *ab initio* molecular dynamics trajectories [2]. Our analysis uncovers a general exponential law governing many-ion correlations, with concerted motion involving ten or more ions being the norm rather than the exception—particularly in fast-conducting Li- and Cu-based systems. Importantly, we demonstrate that such correlated transport is largely temperature-independent and strongly tied to long hop lengths and residence times, calling into question the widespread use of dilute-solution approximations like the Nernst–Einstein relation.

Together, these studies establish a unified theoretical framework for understanding and designing SSEs. By highlighting the indispensable roles of anharmonic lattice vibrations and collective ion dynamics, this work opens new avenues for data-driven discovery and rational optimization of ion-conducting materials.

Exploring Polyanionic Compounds for K-Ion Batteries

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Potassium-ion batteries (KIBs) are attracting attention as a post-lithium and post-sodium technology based on earth-abundant resources, offering a high working voltage owing to the low standard potential of potassium. Their practical use requires positive electrodes that reversibly (de)insert the large K^+ ion at high potential. Open-framework compounds, including polyanionic compounds and metal–organic phosphate open frameworks (MOPOFs), are promising hosts owing to their three-dimensional channels, high voltages from the inductive effect, and structural stability. Here we overview our exploration of potassium insertion hosts, focusing on transition-metal phosphates, MOPOFs, silicates, and fluorides.

Among phosphates and sulfates, the $KTiOPO_4$ -type $KFeSO_4F$ and isostructural $KFePO_4F$, offering large channels for K^+ diffusion, showed reversible Fe^{3+}/Fe^{2+} -based insertion. The MOPOF $K_6(VO_2)(V_2O_3)_2(PO_4)_4(P_2O_7)$ delivered ~ 60 mAh g^{-1} at 4.0 V, and $K_2[(VOHPO_4)_2(C_2O_4)]$ exhibited >100 mAh g^{-1} with excellent rate capability.

In addition to the phosphates and sulfates, transition-metal silicates, K_2FeSiO_4 and K_2CoSiO_4 , prepared by a solid-state route showed reversible one-electron $Fe^{2+}/3^{+}$ and $Co^{2+}/3^{+}$ redox. We further synthesized a complete $K_2Fe_{1-x}Co_xSiO_4$ solid solution, including the first evaluation as potassium insertion hosts, with the average voltage continuously tunable between 2.9 V ($x = 0$) and 3.4 V ($x = 1$). Fe-rich compositions show superior cycling stability, and ex-situ analysis and first-principles calculations reveal that K_2FeSiO_4 exhibited smaller volume change of 1.8% than K_2CoSiO_4 (5.6%).

We will also discuss potassium vanadium fluorides: K_3VF_6 shows a reversible one-electron $V^{3+/4+}$ redox (95 mAh g^{-1} , 3.7 V) and K_2TiF_6 a reversible $Ti^{4+/3+}$ insertion. These frameworks broaden the design space of positive electrodes for potassium-ion batteries.

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Tuning carrier transport for fast charging sulfide based all-solid-state batteries

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The rapid development of lithium-ion battery technology has continuously driven improvements in battery energy density, thereby significantly enhancing the driving range of new energy vehicles. At present, the range of mainstream electric vehicles has exceeded 500 km, yet their charging time typically still exceeds one hour. This has gradually shifted users' concern from "range anxiety" to "charging anxiety." Such a shift imposes higher demands on the fast-charging capability of batteries. However, high-rate charge/discharge processes are prone to safety issues such as lithium dendrite growth and thermal runaway, which are particularly severe in the confined spaces of automobiles. Solid-state lithium batteries, with their superior safety performance, offer a promising path forward; developing solid-state batteries with fast-charging capability is expected to achieve both high performance and high safety in energy storage devices. To this end, starting from the key factors that affect the fast-charging performance of batteries and materials, we propose a series of strategies to enhance the fast-charging capability of solid-state batteries: (i) constructing an integrated macroscopic solid–solid interface through an electrolyte "three-phase percolation" combined with in-situ solidification, enabling smooth ion transport across the interface; (ii) developing electrode materials with both high electronic and ionic conductivity to achieve synergistic and efficient transport of charge carriers; and (iii) proposing a new mechanism of crystalline self-organized hetero-nanodomains to improve the ionic conductivity of electrolyte materials. Based on the above strategies and the developed dry or wet film-forming processes, the prepared sulfide-based solid-state batteries exhibit excellent rate performance while balancing energy density and safety.

Functional coating layer for lithium-sulfur cells with high stability

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Lithium–sulfur cells are promising next-generation energy-storage systems because of their high theoretical energy density (2600 W h kg^{-1}). However, their practical application is hindered by the instability of lithium metal anodes, particularly under lean-lithium conditions required for high-energy-density cells. In this study, we systematically investigate artificial protective interphases based on lithium halides to stabilize thin lithium metal anodes for lithium–sulfur cells. Thin $50 \text{ }\mu\text{m}$ lithium foils are selected to emulate practical low N/P ratio configurations and amplify interfacial degradation. Ex situ protective coatings consisting of lithium halides are fabricated by a simple drop-casting process and comparatively evaluated using lithium//lithium symmetric cells, lithium//copper half cells, and high-sulfur-loading lithium–sulfur full cells. Among the investigated coatings, LiF exhibits the most effective interfacial stabilization, delivering the lowest polarization, the longest cycling stability under various current densities, and suppressed dendritic lithium growth. The superior performance is attributed to the highly stable and polar LiF-rich interface, which promotes uniform lithium-ion transport while mitigating parasitic reactions and irreversible lithium consumption. These findings provide a systematic comparison of lithium halide protective layers and demonstrate the potential of LiF-based artificial interphases for enabling practical, high-energy-density lithium–sulfur batteries with limited lithium excess.

Garnet-solid electrolytes: processing, interfaces, and prospects for solid-state lithium-metal batteries

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Solid-state lithium-metal batteries (SSLMBs) are a promising next-generation energy storage technology, offering the potential for enhanced safety and high energy density. Among solid electrolytes, garnet-type oxides, particularly $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO), stand out due to their good chemical stability against lithium metal, wide electrochemical window, and high ionic conductivity. However, the practical implementation of LLZO faces three critical challenges: difficulty in fabricating thin, large-area membranes, high interfacial resistance with electrode materials, and susceptibility to lithium dendrite growth. In this talk, I will discuss recent advances in garnet electrolytes, emphasizing the shift from high-temperature sintered pellets to low-temperature processed, LLZO-based composite and flexible sheet electrolytes for improved manufacturability. We evaluate strategies for mitigating interfacial issues, including surface cleaning, interlayer engineering, and polymer integration, while also assessing the cycling performance and energy density of LLZO-based pouch cells. Our analysis indicates that despite significant progress, the maximum gravimetric energy density of polycrystalline LLZO-based cells reaches only about 272 Wh kg^{-1} under ideal conditions, highlighting the need for continued research into interface engineering, scalable processing, and dendrite suppression to unlock the full potential of garnet electrolytes.

Mitigating Aluminium Contamination in Garnet Solid Electrolytes via Rational Substrate Design

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All-solid-state lithium batteries (ASSLBs) utilizing garnet-type lithium conductors, such as $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO), represent a highly promising next-generation energy storage technology owing to their high ionic conductivity and electrochemical stability. However, achieving sufficient densification requires high-temperature processing (900-1200°C), which persistently introduces uncontrolled aluminium (Al) contamination from alumina crucibles and processing media, severely altering the electrolytes' structural and electrochemical properties. Small amounts of intentionally introduced Al can stabilize the highly conductive cubic garnet phase, which enhances lithium-ion transport. However, uncontrolled incorporation leads to local compositional heterogeneity, the formation of secondary phases, grain boundary modifications, and it makes it difficult to achieve reproducible electrochemical performance. To resolve this two-decade-long challenge, we systematically evaluated the interfacial reactivity of two representative garnet electrolytes, $\text{Li}_5\text{La}_3\text{Ta}_2\text{O}_{12}$ (LLTO) and $\text{Li}_{6.6}\text{La}_3\text{Zr}_{1.6}\text{Ta}_{0.4}\text{O}_{12}$ (LLZTO) (LLZTO), against oxide substrates with distinct Al environments, including $\alpha\text{-Al}_2\text{O}_3$ (alumina), $\text{Y}_3\text{Al}_5\text{O}_{12}$ (YAG), YAlO_3 (YAO), LaAlO_3 (LAO). Using a combination of experimental methods (accelerated equimolar compatibility studies, solid-state NMR, XPS depth profiling, FIB-SEM, in-situ high-temperature X-ray diffraction) and multiscale computational techniques (first-principles density functional theory calculations, machine learning Molecular Dynamics and continuous modelling), we mapped the relationship between substrate crystal chemistry and garnet phase stability. Our findings demonstrate that while conventional AO and YAG substrates trigger extensive Al contamination and structural decomposition, LAO exhibits exceptional chemical compatibility. Spectroscopic, microscopic, and computational analyses reveal that Al remains largely confined to its stable octahedral sites within the LAO perovskite structure, making Al incorporation thermodynamically endothermic at typical sintering temperatures and restricting its transport to a narrow interaction zone. Ultimately, this work demonstrates that substrate-induced degradation can be effectively mitigated through rational substrate design and interfacial engineering rather than the complete elimination of Al-containing crucible materials.

Interfacial Regulation of Lithium Metal Anodes via Nitrate-Induced Lithiophilic Frameworks for Ultralow Overpotential Alkali Metal Batteries

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Lithium metal anodes are widely regarded as a key enabler for next-generation alkali metal batteries, yet their practical deployment remains hindered by unstable solid electrolyte interphases and uncontrolled dendritic growth. In this work, we present a nitrate-enabled interfacial engineering strategy based on a binder-free graphene oxide (GO) framework modified with silver nitrate (AgNO_3), which simultaneously regulates interphase chemistry and lithium nucleation behavior. Upon initial lithiation, the nitrate species promote the formation of Li_3N -rich interphases, while Ag^+ is reduced to generate lithiophilic Ag seeds that guide uniform lithium deposition. This dual-function design establishes a chemically and structurally stable interface, leading to significantly reduced interfacial resistance and prolonged cycling stability. Symmetric $\text{Li}||\text{Li}$ cells exhibit ultralow overpotentials and extended lifetimes over thousands of cycles under practical current densities. Comprehensive characterizations, including X-ray photoelectron spectroscopy with depth profiling and electron microscopy, reveal the dynamic evolution of the interphase and confirm the formation of inorganic-rich, compact SEI layers. The results highlight the importance of coupling interphase chemistry with nucleation control to achieve stable lithium metal operation. This work provides insights into rational interfacial design for alkali metal batteries and offers a scalable strategy for improving lithium metal anodes. We believe such approaches open new opportunities for collaboration between academia and industry, particularly in advancing practical high-energy-density battery systems and bridging fundamental understanding with manufacturable technologies.

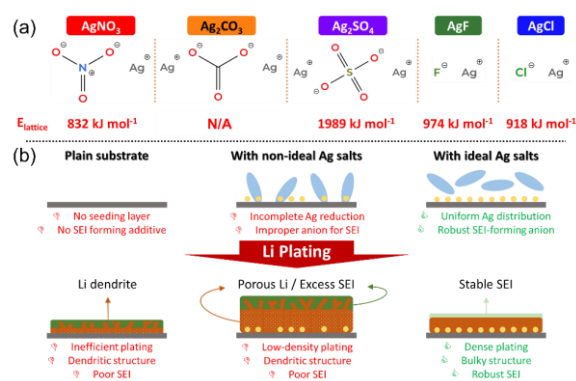


Figure 1. (a) Lattice energy-dependent dissociation behavior of silver salts and its influence on ionic availability in the electrolyte. (b) Correlation between salt dissociation, Ag nucleation behavior, and anion-derived SEI chemistry governing lithium deposition.

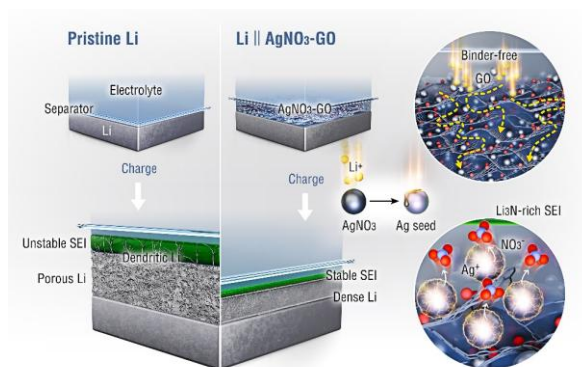


Figure 2. AgNO₃-confined graphene oxide (SN-GO) framework enabling coupled regulation of Ag nucleation and Li₃N-rich SEI formation for stable lithium deposition.

Beyond 99.9% Coulombic Efficiency in Aqueous Zinc Metal Batteries

Without Relying on SEI

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Aqueous rechargeable batteries offer high intrinsic safety, relatively low material and manufacturing costs, and straightforward scalability. However, even after several decades of extensive research, their practical implementation remains limited by persistent difficulties in maintaining stable electrochemical operation over prolonged cycling.

This presentation examines the fundamental and practical factors governing anode instability, with particular emphasis on aqueous Zn-metal batteries. The discussion begins with an experimentally derived potential diagram for Zn plating/stripping and hydrogen evolution, which provides a direct framework for evaluating the electrochemical stability of Zn anodes. In particular, the electrostatic environment surrounding Zn^{2+} can shift both the Zn^{2+}/Zn redox potential and the hydrogen evolution potential. Rational control of these potential shifts can enable highly reversible Zn plating/stripping with Coulombic efficiencies exceeding 99.9% and provide rational guidance for the design of future aqueous electrolytes. The presentation also discusses the intrinsic difficulty of forming a Zn^{2+} -conducting SEI and maintaining its protective function, together with the remaining uncertainties in mechanistic interpretation and electrochemical evaluation.

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Hydration thermodynamics of $\text{Na}_2\text{Fe}_2(\text{SO}_4)_3$ defines its ambient storage and closed-loop recycling conditions

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Alluaudite-type sodium iron sulfate, $\text{Na}_{2.5}\text{Fe}_{1.75}(\text{SO}_4)_3$ is a promising low-cost, 3.8V cathode material currently discussed for application in mass-production sodium-ion batteries. However, its practical deployment has been constrained by its hygroscopic nature, which complicates manufacturing and storage. Here, we resolve the hydration behaviour of $\text{Na}_{2.5}\text{Fe}_{1.75}(\text{SO}_4)_3$ as a function of water vapor pressure ($P_{\text{H}_2\text{O}}$) and temperature, identifying a well-defined hydration boundary and deliquescence threshold that separates anhydrous, hydrated, and aqueous phase regimes. These boundaries define the practical storage stability window in which anhydrous $\text{Na}_{2.5}\text{Fe}_{1.75}(\text{SO}_4)_3$ remains stable over extended timescales. Our results further show that hydration proceeds via discrete crystalline phase transitions rather than continuous degradation, enabling moisture sensitivity to be treated as a thermodynamically governed property. By leveraging this behaviour, we demonstrate a method for closed-loop aqueous-based recycling of degraded $\text{Na}_{2.5}\text{Fe}_{1.75}(\text{SO}_4)_3$ material. This recycling process achieves high cathode recovery (~96.82%wt.) of the $\text{Na}_{2.5}\text{Fe}_{1.75}(\text{SO}_4)_3$ phase with electrochemical performance equivalent to the pristine material across 200 cycles. Life cycle and technoeconomic analysis indicate recycled $\text{Na}_{2.5}\text{Fe}_{1.75}(\text{SO}_4)_3$ exhibits substantially reduced environmental impact and cost relative to established battery cathode chemistries, including Lead–Acid, LiFePO_4 (LFP), and Ni–Co–Mn (NCM622). These results position NFS as a low-impact, economically favourable cathode material and recast its moisture sensitivity not as a limitation, but as an enabling feature for circular design and aqueous reprocessing of next-generation battery cathode materials.

Understanding the Materials and Chemistry in Rechargeable Li(Na)/Cl Batteries

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Lithium-thionyl chloride primary battery is a widely deployed commercial battery, which exhibits outstanding electrochemical performance under extreme environments, including an ultra-wide operating temperature range, high energy density, and long shelf-life. Investigations into the materials and chemistry of this battery system may enable its secondary-battery transformation, facilitating the development of a new class of rechargeable batteries, namely lithium-chlorine secondary batteries. This report presents our recent research findings. Focusing on the influences of electrode materials and electrolytes on electrochemical processes in lithium (sodium)-chlorine secondary batteries, we explore the correlation between electrode material structure and performance, as well as the functional mechanism of electrolytes in the corresponding electrochemical reactions. Optimizing the structure and composition of electrode materials together with the solvation behavior of electrolytes can effectively improve the electrochemical performance and cycle life of such batteries, which further contributes to the fundamental understanding and research of this emerging rechargeable battery system.

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Behavior of Sodium Metal Electrode and Some Effective Additives

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Sodium metal has shown impressive potential as a negative electrode material for high energy density sodium metal batteries on the grounds of its high specific capacity (1166 mAh g^{-1} based on the weight in the charged state) and low potential (-2.71 V vs. SHE). It provides completely different behaviors, depending on the electrolytes.[1,2] In this work, recent progress of the sodium metal chemistry in our group will be presented. While high sodium metal plating-stripping efficiency has been reported in ether-based electrolytes, we usually use sodium metal counter electrodes with different electrolytes in half-cell tests. Our detailed investigation revealed that polarization of Na metal electrode significantly varies depending on the electrolytes and also additives, and thus the half-cell tests using the sodium metal counter electrode are not trustable enough to assess the target electrode. Instead, we propose the NASICON-type electrode materials as a counter electrode as they give flat potential profile with small polarization.[3]

A range of additives based on fluorooxalato-complex anions were investigated to improve the performance of sodium-ion batteries. Our DFT calculations showed the anodic and cathodic unstabilities of the oxalate ligand compared to fluoride ligand, which contributes to the formation of solid-electrolyte interphase and cathode-electrolyte interphase. Appropriate use of additives improved rate, coulombic efficiency, and cycle stability of the full cells.

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Polymer-based solid-state electrolytes

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Solid-state lithium metal batteries are widely regarded as the next-generation direction for battery development, with solid-state electrolytes serving as their core material. Compared with inorganic solid-state electrolytes, polymer-based solid-state electrolytes offer clear advantages in terms of thinning, scalability, and cost. However, conventional polymers suffer from shortcomings such as low room-temperature ionic conductivity, low lithium-ion transference number, and poor high-voltage tolerance. High lithium-ion conduction and ultra-thinning represent the development trends for polymer-based solid-state electrolytes. Our team has previously innovatively designed copolymer molecular structures, functional fillers, support membrane pore structures, and the electrode/electrolyte solid-solid interface. The as-prepared polymer solid-state electrolyte membranes have tunable thicknesses in the range of 3-20 μm , a room-temperature ionic conductivity reaching $0.75 \text{ mS} \cdot \text{cm}^{-1}$, a lithium-ion transference number exceeding 0.8, and a wide electrochemical work window ($\geq 5 \text{ V}$). All-solid-state pouch cells assembled with these membranes, high-nickel ternary cathodes, and lithium metal anodes exhibit excellent tolerance to bending and shearing, and can operate normally at temperatures as low as below -15°C . This study provides a reference for designing polymer-based solid-state electrolytes with comprehensively superior performance.

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Balancing ion transport, high-voltage stability and sustainable design in solvent-free UV-crosslinked solid polymer electrolytes

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Commercial alkali metal-ion batteries rely on liquid electrolytes containing volatile organic carbonate solvents, which raise safety concerns due to side reactions, gas evolution, and flammability. All-solid-state batteries based on polymer electrolytes represent a promising alternative, offering improved safety, mechanical flexibility, and compatibility with next-generation battery architectures¹.

This contribution presents recent advances in high-performance polymer electrolytes for solid-state lithium- and sodium-based batteries. Particular emphasis is placed on solvent-free fabrication strategies, including UV-induced photopolymerization and hybrid polymer electrolytes incorporating room-temperature ionic liquids, low-volatility additives, and sustainable materials. These approaches enable enhanced ionic transport, improved interfacial stability, and stable electrochemical performance under ambient operating conditions^{2,3}.

To further address sustainability challenges, bio-derived and recycled polymers, including polyhydroxyalkanoates (PHAs) and recycled poly(vinyl butyral) (PVB), have been incorporated into polyethylene oxide (PEO)-based electrolyte systems and battery components. The resulting materials exhibit improved mechanical robustness, thermal stability, and electrochemical performance while promoting the use of renewable and waste-derived resources⁴. Their applicability has been demonstrated not only as solid electrolytes but also as sustainable binders and separators⁵.

The technological relevance of the developed materials was assessed through both laboratory-scale and industrial-format full cells. These results demonstrate the potential of sustainable polymer engineering to simultaneously address performance, safety, and environmental challenges, providing a realistic pathway toward next-generation lithium- and sodium-based energy storage technologies.

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Designing High-Deformability Materials for All-Solid-State Batteries Through Shear Modulus Engineering

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Weak contact and high resistance between ceramic powder particles limit their applicability in all-solid-state Li-ion batteries, particularly those assembled via powder moulding. To overcome these limitations, sulfide and chloride materials have been extensively studied because their deformability and Li diffusivity are typically greater than those of oxide materials. However, not all sulfide and chloride materials exhibit the high Li diffusivity and deformability necessary for all-solid-state batteries. This study focused on determining the suitability of the shear modulus as an index of deformability, in addition to a Li diffusivity index. These indices were calculated for all the Li-containing chloride compounds in a structural database. Six chloride materials (Li_2CoCl_4 , Li_2CrCl_4 , $\text{Li}_{10}\text{Mg}_7\text{Cl}_{24}$, $\text{Li}_4\text{Mn}_3\text{Cl}_{10}$, Li_2FeCl_4 , and LiAlCl_4) with different shear moduli were experimentally evaluated. The Li diffusion coefficients of most of these chlorides were found to be higher than those of the oxide and sulfide electrode materials typically employed in Li-ion batteries. Moreover, deformability, which for compressed pellets includes contributions from the relative density and grain boundary resistance, was observed to vary among these chlorides. The shear modulus, as determined by first-principles calculations, was confirmed to be a suitable screening index for deformability. The utilisation of the design guidelines for deformability is expected to facilitate significant advancements in all-solid-state battery material research.

Moisture-Assisted Hydroflux Synthesis of Layered Cathodes

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Layered cathodes, such as LiCoO_2 (LCO) and LiNiO_2 (LNO), are usually synthesized at high temperatures ($\geq 700\text{ }^\circ\text{C}$) via a conventional solid-state process. We recently proposed an effective synthesis approach, the “hydroflux process,” to produce a highly crystalline layered LCO at low temperatures ($\leq 300\text{ }^\circ\text{C}$) in ≤ 1 h. A series of XRD studies revealed that layered LiCoO_2 forms even at $150\text{ }^\circ\text{C}$. We also confirmed that the water molecules in the molten mixed hydroxides significantly accelerate the crystal growth of layered LCO. Under strongly alkaline conditions, the soluble HCoO_2^- anion species enable fast, low-temperature crystal growth of LiCoO_2 without the formation of the spinel Co_3O_4 . In situ XAS studies also revealed that the moisture-assisted atmosphere induces a local structural transformation of $\text{Co}(\text{OH})_2$ toward an $\alpha\text{-Co}(\text{OH})_2$ -like coordination environment at room temperature. Layered LCO formation mostly completes at $150\text{ }^\circ\text{C}$. The continuous crystal growth of LCO above these temperatures indicates the existence of Co^{3+} soluble species under hydroflux conditions.

The moisture-assisted synthesis of $\text{LiNi}_{1/3}\text{Mn}_{1/3}\text{Co}_{1/3}\text{O}_2$ (NMC111) and other NMC-type cathodes was also performed. Although NMC111 is readily obtained at $300\text{ }^\circ\text{C}$, cation mixing between Li and Ni remains a problem because Ni^{3+} species can easily be reduced to Ni^{2+} by water molecules. Therefore, high-temperature calcination is still necessary to obtain high-quality NMC-type cathodes. We also propose a two-step heating approach as an effective method for synthesizing NMC-type cathodes. In the first step, heat the precursor materials under an inert, moisture-free atmosphere to promote crystal growth; subsequently, calcine the sample under a dry O_2 atmosphere to oxidize the transition metals. The two-step heating process enables us to synthesize NMC-type cathodes even without using the transition-metal mixed hydroxide synthesized by co-precipitation as a starting material.

Defect-engineered Oxide-based Electrode Materials for Li Storage

Applications

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Ni-rich layered oxides such as LiNiO_2 are attractive positive electrodes, but suffer from capacity degradation at high voltages due to surface destabilization. Recent studies show that non-stoichiometry and antisite defects play a critical role, and defect engineering enables highly reversible pure Ni-based layered materials without metal substitution.² In parallel, Co-/Ni-free Mn-based electrodes are important for cost-effective electric vehicles.²⁻⁵ While typically synthesized by energy-intensive milling, nanostructured LiMnO_2 with high energy density ($\sim 800 \text{ W h kg}^{-1}$) has recently been obtained through conventional calcination, demonstrating scalability. For safety, solid electrolytes are promising, but electrode volume changes complicate interface stability. Dimensionally invariable cation-disordered rocksalt oxides address this issue, achieving excellent reversibility with solid electrolytes.^{6,7}

Overall, defect-engineered and nanostructured lithium insertion materials are central to the development of safe, high-energy, and practical Li-ion batteries.

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NFPP Cathodes for Sodium-Ion Batteries: From Material Design to Scale-Up

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Sodium-ion batteries are emerging as a cost-effective and sustainable complement to lithium-ion technology. Among cathode materials, $\text{Na}_4\text{Fe}_3(\text{PO}_4)_2\text{P}_2\text{O}_7$ (NFPP) combines an earth-abundant composition with a robust polyanionic framework, good air stability, and a theoretical capacity of 129 mAh g⁻¹ at an average potential of approximately 3.0 V vs. Na⁺/Na. Its practical performance nevertheless depends on controlling phase formation, carbon coating, and electrode processing.

This invited talk presents our work on NFPP from material optimization to synthesis scale-up and cell-level validation. Conventional solid-state synthesis can generate secondary phases, particularly electrochemically inactive maricite NaFePO_4 and $\text{Na}_2\text{FeP}_2\text{O}_7$.¹ Controlled sodium and phosphorus excess was investigated to steer phase formation, limit unwanted products, and improve electrochemical utilization. In parallel, the carbon precursor and thermal treatment strongly affect carbon distribution, electronic conductivity, particle morphology, rate capability, and cycling stability.² The results show that precursor stoichiometry and carbon coating must be optimized together.

Current activities address transfer to larger and more reproducible batches through adapted precursor preparation and thermal processing, including spray-drying-based routes aimed at improving homogeneity and facilitating scale-up. The powders are processed into water-based electrodes at practically relevant mass loadings using standard slurry mixing, coating, drying, and calendering. Evaluation in half cells and hard-carbon full cells links powder properties to electrode- and cell-level performance. Optimized NFPP has shown capacity retention of up to 98% after 500 cycles under the investigated conditions. Overall, the work highlights the need to consider composition, synthesis, coating, and electrode design as one continuous development chain to translate the intrinsic advantages of NFPP into scalable sodium-ion battery cells.

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Utilization of Cationic Multi-electron Redox in Na-Superrich Oxides for Sodium-Ion Battery

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Iron and manganese oxide-based materials are attractive cathodes due to their earth-abundant nature. Especially in iron oxide materials, there have been efforts made for a long time to utilize $\text{Fe}^{4+}/\text{Fe}^{3+}$ redox reactions at cathodes. Even in well-known layered-type oxides such as LiFeO_2 and NaFeO_2 , the utilization of $\text{Fe}^{4+}/\text{Fe}^{3+}$ one-electron redox is still challenging; excess oxidation leads to O_2 evolution. Utilization of anionic oxygen redox in solids is a well-known strategy to achieve large reversible capacity, while large voltage hysteresis due to instability at the charged phase, i.e., the oxidized oxygen species, remains a critical issue.

Herein, we introduce a new material of Na-Fe binary oxides, Na_5FeO_4 , a thermodynamically stable phase in the Na-Fe-O phase diagram, for a sodium-ion battery cathode. The sodium-superrich Na_5FeO_4 is composed of tetrahedral FeO_4 units, which is different from NaFeO_2 (composed of octahedral FeO_6 units). It delivers a two-electron redox reaction to achieve a high reversible capacity of 228 mAh g^{-1} . X-ray absorption spectroscopy suggests that during the two-electron redox, a cationic $\text{Fe}^{5+}/\text{Fe}^{3+}$ redox with no O_2 evolution proceeds, holding the tetrahedral FeO_4 local structure.

Na-Layer Chemistry and Cathode Properties of P2/P3-Type Layered Oxides

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Among various cathode materials for sodium-ion batteries (SIBs), P-type layered transition metal oxides are considered one of the most promising candidates owing to their open Na-layer framework, which facilitates Na⁺ diffusion and enables high-power performance. In these materials, the Na-layer environment is governed by several structural factors, including the oxygen stacking sequence, Na-site coordination, interlayer spacing, and local chemical environment. These factors strongly influence Na-ion transport, structural stability, phase-transition behavior, and ultimately electrochemical performance.

In this presentation, recent studies on P-type layered oxide cathodes are presented with a focus on the relationship between the Na-layer environment and electrochemical properties through two distinct approaches as follows:

1) Effect of Na-layer stacking sequence on electrochemical properties: Highly crystalline P3-type Na_{2/3}Ni_{1/3}Mn_{2/3}O₂ was synthesized via a spinel-derived route, enabling a direct comparison with its P2 counterpart under comparable conditions. By minimizing the influence of crystallinity, the intrinsic effect of the Na-layer stacking sequence on electrochemical behavior was clarified.^{1,2)}

2) Systematic comparison of alkaline earth metal doping effects on the Na-layer: The effects of alkaline earth metal dopants on P2-type Na_{2/3}Fe_{1/2}Mn_{1/2}O₂ were systematically compared, with particular emphasis on the Na-layer environment. Partial substitution at the Na sites expands the interlayer spacing and modifies the local Na coordination environment, leading to enhanced structural stability and Na-ion transport at some cases.^{3,4)}

As a results, these studies demonstrate that the Na-layer environment plays a crucial role in determining the electrochemical properties of P-type layered oxide cathodes.

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Enhancing Oxygen Redox Kinetics in Na-Layered Cathodes toward High-Power and High-Energy Sodium-Ion Batteries

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In this study, we investigate Na-layered cathode materials designed to overcome these limitations and enable stable oxygen redox behavior even under fast discharging conditions. By optimizing the local structure and electronic environment, we suppress structural instabilities typically associated with oxygen activity and improve the overall reaction kinetics. The resulting materials demonstrate both high energy density and excellent fast discharging performance, with minimal voltage decay over prolonged cycling. Our findings provide insight into how oxygen redox can be harnessed more effectively in Na-layered oxides and point toward practical strategies for advancing high-performance SIB cathodes suitable for demanding energy storage applications.

Sustainable Chemical Delithiation of LiFePO_4 and Its Reuse for Sodium Batteries

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Triphylite NaFePO_4 , an olivine-type cathode material composed of earth-abundant elements, is a promising candidate for sodium-ion batteries owing to its high capacity and energy density [1,2]. However, its direct synthesis remains challenging because the thermodynamically metastable triphylite phase readily transforms into the electrochemically inactive maricite phase [3].

In this study, we present a sustainable route for producing heterosite FePO_4 , the fully delithiated form of LiFePO_4 , through O_2 -driven chemical delithiation of LiFePO_4 in an aqueous acetic acid solution. The process operates in a closed-loop system, where oxygen serves as the oxidant and lithium is recovered as lithium acetate for reuse, enabling cost-effective and circular FePO_4 production. Furthermore, we propose a lean sodium-metal battery configuration consisting of a thin sodium-metal anode and FePO_4 positive electrode. This design improves cycling stability while maintaining high energy density, providing a practical strategy for next-generation sodium battery systems [4].

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Stabilizing Oxygen Redox in Sodium Manganese Oxide Cathodes through Doping and Cooling Engineering

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Oxygen redox in manganese oxide cathodes offers a new pathway to exceed the capacity of conventional cationic redox of sodium-ion batteries in the high-voltage region. However, its practical application is hindered by cation migration, oxygen loss, and capacity fading. In this work, Li/F co-doping and tailored synthesis conditions were implemented to improve the oxygen redox reversibility. $\text{Na}_{0.7}\text{Li}_{0.1}\text{Mg}_{0.15}\text{Mn}_{0.75}\text{O}_{1.9}\text{F}_{0.1}$ (NLMMOF) was designed to demonstrate that Li substitution activates oxygen redox and F substitution strengthens the metal-anion bonding. In addition, slow cooling and quenching after the synthesis of $\text{Na}_2\text{Mn}_3\text{O}_7$ (NMO) were conducted to elucidate how the cooling process can regulate the structural motif of Mn vacancy ordering. In-situ XRD, ex-situ EXAFS, and in-situ DEMS collectively reveal that the improved oxygen redox stability of NLMMOF originates from suppressed P2-O2 phase transition, minimal local structural distortion, and negligible oxygen evolution. Time-resolved XRD, high-resolution TEM, and in-situ DEMS analyses comprehensively reveal that the enhanced oxygen redox reversibility of slow-cooled NMO arises from a homogeneous stacking-fault-rich architecture, mitigated Mn in-plane migration, and suppressed O_2 release. These co-doping and cooling engineering strategies provide a practical design principle for stabilizing oxygen redox in layered oxide cathodes for advanced sodium-ion batteries.

Keywords: oxygen redox, oxide cathodes, sodium-ion batteries, co-doping, slow cooling

Nanostructured Sodium Vanadium Phosphate-Based Cathode Materials for Sodium-Ion Rechargeable Batteries

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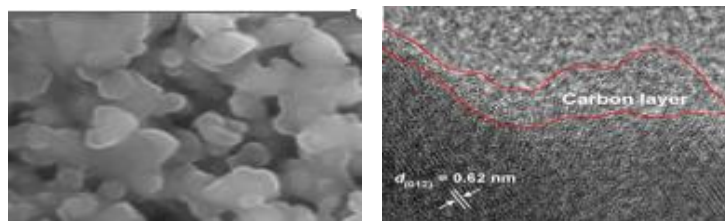
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The growing demand for sustainable energy storage, driven by climate concerns and the rapid adoption of renewable energy sources, has intensified research into alternatives to lithium-ion batteries (LIBs). Although LIBs dominate the market, their large-scale use is limited by lithium scarcity, uneven geographical distribution, and increasing cost. In this regard, sodium-ion batteries (SIBs) have emerged as a promising low-cost and sustainable option due to the abundance and wide availability of sodium resources. Among cathode materials for SIBs, $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ (NVP) has attracted considerable attention because of its NASICON-type three-dimensional framework, which provides fast Na^+ ion diffusion and excellent structural stability. However, its poor intrinsic electronic conductivity restricts high-rate performance. This work focuses on enhancing the electrochemical behaviour of carbon-coated NVP (NVP@C) through controlled synthesis, nanostructuring, and thermal optimization using sol-gel and hydrothermal methods. For sol-gel synthesized samples, the optimized NVP@C calcined at 900 °C crystallized in a rhombohedral phase with porous interconnected morphology and uniform carbon coating. It delivered an initial discharge capacity of 99 mAh g^{-1} at 1C, retained 73 mAh g^{-1} at 10C, and exhibited 93% capacity retention after 1400 cycles at 10C. A symmetric full-cell based on this material showed 80% capacity retention over 500 cycles at 2C.

For hydrothermally synthesized samples, Rietveld refinement confirmed phase purity and temperature-dependent structural variations. The optimized sample calcined at 850 °C delivered 94 mAh g^{-1} with 90% capacity retention over 250 cycles at 2C and retained 63 mAh g^{-1} after 2000 cycles at 10C. A practical sodium-ion full cell using optimized NVP@C cathode and a hard carbon anode was also demonstrated. With an optimized N/P ratio of 0.75, the device delivered ~60 mAh g^{-1} initially and retained ~55 mAh g^{-1} after 50 cycles. These results highlight the strong potential of optimized NVP cathodes for next-generation high-performance SIBs.

Keywords: $\text{Na}_3\text{V}_2(\text{PO}_4)_3$ cathode, Hard carbon anode, Sodium-ion full cell, N/P ratio, Cycling stability



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Optimized Hard Carbon for High-Performance Sodium-Ion Batteries

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Hard carbon (HC) has emerged as one of the most promising anode materials for sodium-ion batteries because of its affordability, long-term cycling stability, and favorable sodium storage capability. Nevertheless, its practical implementation in full-cell systems is hindered by limited reversible capacity and relatively low initial Coulombic efficiency (ICE). In this work, the synthesis strategy was systematically optimized by tailoring the carbonization and pre-treatment processes to enhance the structural and electrochemical properties of HC. Increasing the carbonization temperature promoted a higher degree of graphitization, while the influence of pre-oxidation conditions and heating rate on material characteristics was comprehensively investigated. Structural analyses revealed that the optimized HC possessed enlarged graphitic domains, reduced specific surface area, and lower porosity, all of which contributed to improved sodium storage performance. The optimized electrode exhibited a reversible capacity of 320 mAh g⁻¹ together with excellent cycling stability in sodium half-cells. Furthermore, pre-oxidation was found to increase the concentration of carbonyl (C=O) functional groups, providing additional active sites for sodium-ion adsorption. Fine-tuning the heating ramp rate further enhanced the electrochemical performance, demonstrating that careful control of both structural ordering and surface chemistry is an effective approach for improving the capacity and efficiency of hard carbon anodes for advanced sodium-ion batteries

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Beyond Graphite: Na⁺ - Solvent Co-Intercalation in Organic and Carbon Hosts

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The rise of sodium-ion battery research has been closely linked to the search for reliable negative electrode hosts, as sodium does not form intercalation compounds with graphite in the same manner as lithium. In contrast, disordered carbons or solvent co-intercalation mechanism enable reversible sodium storage.

In the current presentation, we report recent findings on electrochemical sodium storage in materials with a lower degree of structural order than graphite, including chemical vapor deposition (CVD)-derived carbons and organic synthesis-derived conjugated polyarenes. Despite different structures, both classes of materials undergo reversible Na⁺-solvent co-intercalation in ether-based electrolytes, resulting in fast reaction kinetics and favorable rate capability.

The electrochemical mechanism is investigated via *in situ* Raman spectroscopy complemented by *in situ* EPR for conjugated polyarenes, enabling direct observation of structural and electronic changes during reversible (de)sodiation. These findings are compared with the well-established behavior of graphite and hard carbon to identify similarities and differences in their sodium storage mechanisms. Finally, the influence of material structure, synthesis conditions, and electrolyte composition on the electrochemical performance and reaction pathway is discussed.

Electrochemical activation of V^{3+} and V^{4+} oxides in aqueous zinc-ion batteries

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Abstract. Rechargeable aqueous zinc-ion batteries (RAZIBs) are recognized as promising energy storage systems to meet the ever-growing demand for grid-scale applications. Developing reliable cathode materials with superior electrochemical performance plays a decisive role in this field. Electrochemical activation (ECA) has been recognized as an appealing strategy to enhance the performance of vanadium oxides. In this presentation, we reveal that spinel ZnV_2O_4 and tetragonal VO_2 can be electrochemically activated by applying anodic potentials, and the upper potential limit (UPL) plays a decisive role in affecting battery performance. During the activation process, the parent vanadium oxides are found to undergo water-driven irreversible structural transitions resulting in the formation of amorphous VO_x for charge storage. Nonetheless, treating the sample with high UPLs inevitably exacerbates vanadium dissolution and induces the formation of crystalline $Zn_3(OH)_2V_2O_7 \cdot 2H_2O$ (ZVOH) as an inactive byproduct, thereby causing significant capacity fading during long-term cycling. As such, the electrochemical performance of the samples can be optimized by controlling the UPL. For the activated tetragonal VO_2 , high attainable capacity (353 mAh g^{-1}) and good cycle stability (86% after the subsequent 300 cycles) can be simultaneously achieved. This work sheds light on activating low-valent vanadium-based oxides for practical application in RAZIBs, opening an avenue for developing cathode materials for aqueous batteries.

Recycling, Recovery, and Reuse of High-Purity V₂O₅ Cathode

Materials from Waste Aqueous Zinc-Ion Batteries

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The sustainable deployment of aqueous zinc-ion batteries (AZIBs) requires not only high-performance electrode materials but also practical strategies for recycling and recovery resource-intensive vanadium cathodes. Here, we report a hydrometallurgical route that converts spent AZIB cathodes into reusable vanadium pentoxide (V₂O₅) cathode material. The process integrates acid leaching, selective solvent extraction, stripping, ammonium metavanadate precipitation, and calcination. The effects of extraction pH, reagent concentration, organic-to-aqueous phase ratio, and reaction time were systematically evaluated to improve vanadium separation and product quality. Structural and compositional analyses by SEM–EDS, XRD, and ICP–MS showed that the regenerated V₂O₅ retained a rod-like morphology similar to commercial V₂O₅ and achieved a purity above 99%, with only trace Zn, Ca, K, and Al impurities. To determine whether the recovered product could function beyond a purified intermediate, it was directly fabricated into AZIB cathodes. The regenerated V₂O₅ exhibited rate capability, cycling stability, and electrochemical impedance behavior comparable to those of commercial V₂O₅. Life cycle assessment further indicated environmental advantages relative to primary V₂O₅ production. These results establish a material-to-material regeneration pathway that couples selective vanadium recovery with cathode performance validation, thereby supporting circular material use in aqueous battery systems.

Interfacial Engineering of MnO₂ Cathodes via MOF-5 Thin-Film Modification for Ultra-stable Aqueous Zinc-Ion Batteries

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To address the escalating energy crisis and frequent extreme weather events, it would be crucial to develop energy systems that are reliable, affordable, and environmentally friendly. Aqueous zinc-ion batteries (AZIBs) are emerging as promising future energy storage solutions, thanks to zinc's abundant resources, safety, and low cost. A major focus in AZIB advancement is enhancing electrochemical stability through interfacial engineering. This research presents a new interfacial phase reconstruction technique that utilizes the confined induction properties of MOF-5. This approach involved preloading Mn²⁺ ions into an electrolyte containing 2 M ZnSO₄ and 0.2 M MnSO₄, which primarily interacted with Zn²⁺ ions on the MOF-5 surface. This interaction resulted in a dynamic, reversible in situ ZnMn₂O₄ interphase that significantly improves the performance and stability of AZIBs. Incorporating MOF-5 created confined nucleation sites, encouraging the selective deposition of ZnMn₂O₄ and preventing the collapse and dissolution of the underlying MnO₂. Through this dynamic interfacial engineering, the MOF-5-modified MnO₂ electrode achieved a reversible capacity of 173.7 mAh g⁻¹ at 0.1 A g⁻¹, which decreased to 79 mAh g⁻¹ after 5000 cycles at 1 A g⁻¹. The system could provide notable stability, high reversibility, and low production costs, positioning it as a viable candidate for next-generation energy storage solutions.

Fast Lithium-ion Intercalation in a Mutual Crystalline-Amorphous Single Phase

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Amorphous materials have received much interest due to their complexity and unique chemical structure and properties. In contrast to crystalline phases that show translation symmetry on nanometer to micrometer length scales, amorphous materials lack such long-range order while retaining local order at atomic distances. Efforts to elucidate the structure of 3-dimensional amorphous materials usually lead to the question of whether the particular amorphous phase shows medium-range order, a fundamental consideration in the long-lasting discussion on the gap between amorphous and crystalline materials.

Here we demonstrate a new form of organized inorganic matter that is amorphous in two dimensions, while exhibiting long-range order and a high degree of crystallinity in the third. The structure consists of periodically stacked 2-dimensional amorphous Nb-W-O monolayers without long-range in-plane order. The Nb-W-O nanorod thin films were grown by pulsed laser deposition on conducting Nb-doped single-crystalline SrTiO₃ substrates with (001), (110), and (111) orientations. Detailed reciprocal space mapping and high-resolution scanning transmission electron microscopy was performed to identify the unique structure. Our findings show that the gap between crystalline and amorphous materials does not only depend on medium-range order but can also apply to principal dimensions within the same solid [1].

Fast-charging is one of the key requirements for battery applications, and various niobium-oxide based electrode materials have shown very promising high-rate performance owing to their stable host structure for lithium-ion intercalation. Here we will show the fast-charging performance of these unique single phase solids with orientation-dependent mutual crystalline and amorphous order.

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Addressing key challenges in the development of Zn-based electrochemical storage systems

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Renewable energy production is characterized by intermittent power output and requires large-scale applications to improve energy storage capability (currently, less than 1% of electrical energy is stored). Developing low-cost, environmentally friendly, high-performance electrochemical storage systems is fundamental to a sustainable energy economy. The most mature battery technology is lithium-ion, which is considered one of the most appealing candidates for electric vehicle power sources. However, the large-scale application of lithium-ion batteries is currently under discussion due to the limited lithium and certain transition metals, such as Co and Ni resources. Several other metallic anodic materials such as sodium, potassium, calcium, magnesium, and aluminum [1-3], characterized by a higher abundance of lithium, have been considered suitable candidates for electrochemical storage devices in replacing lithium systems. Notably, significant efforts are being devoted to understanding and addressing key challenges in developing these so-called "beyond Li-ion" chemistries, and substantial insights into their storage and failure mechanisms have been obtained over the past few years through advanced characterization and computational/modeling techniques. We will present our work on developing advanced electrolyte systems for Zn-ion batteries, including concentrated electrolytes and Gel Polymer Electrolytes (GPEs). The electrolyte formulation is based on natural biopolymers and low-cost, sustainable chemicals such as H_2SO_4 , ZnSO_4 , and H_3PO_4 . Particular attention is also paid to regulating Zn ion conduction/transport across the interfaces by adjusting the amount of water and the polymer crosslinking density, coupled with a mechanistic understanding. [4–8]

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Multiple Pathways for Aluminum-Based Energy Storage across Chemistries and Scales

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Aqueous aluminum batteries have emerged as promising candidates for safe and sustainable energy-storage systems because of the high abundance, low cost, and intrinsic safety of aluminum. However, the electrochemical behavior of aluminum anodes in aqueous electrolytes remains controversial, particularly regarding the reversibility of aluminum deposition and the origin of their apparent charge – discharge performance. Addressing these fundamental questions is essential for developing practical aluminum-based electrochemical systems. In this presentation, we first revisit the reaction mechanisms of aluminum and aluminum-alloy anodes in aqueous electrolytes through systematic electrochemical and structural investigations. By examining pristine aluminum, surface-modified aluminum, Al – Cu alloys, and Zn – Al alloys in various aluminum-based aqueous electrolytes, we demonstrate that the observed electrochemical responses originate predominantly from corrosion reactions and hydrogen evolution rather than reversible aluminum plating and stripping. The results establish that chloride-induced corrosion, alloy-assisted hydrolysis, and electrolyte chemistry collectively govern the interfacial behavior of aluminum electrodes, providing a unified mechanistic framework for interpreting aqueous aluminum battery reactions. Building upon these mechanistic insights, we further investigate the unique reaction behavior of an Al – Cu alloy in highly concentrated $\text{Al}(\text{OTf})_3$ aqueous electrolytes. Unlike conventional dissolution-dominated corrosion, the Al – Cu alloy exhibits a passivation-driven surface evolution process. Preferential dissolution of α -Al increases the local pH, promoting the growth of an $\text{Al}(\text{OH})_3$ passivation layer that gradually encapsulates the alloy surface. As the passivation layer thickens, it shields the electrode from the applied electric field while facilitating the in situ formation of Cu and Cu_2O through the reduction of dissolved Cu species. This dynamic interfacial reconstruction suppresses continuous corrosion and stabilizes the electrode surface, revealing an alternative pathway for improving aluminum-based electrodes through controlled passivation rather than reversible aluminum deposition. Together, these studies establish a comprehensive understanding of aqueous aluminum electrochemistry by linking corrosion mechanisms with interfacial evolution. Rather than pursuing reversible aluminum plating under aqueous conditions, the results demonstrate that rational control of corrosion and passivation provides a more realistic strategy for developing durable aluminum-based electrochemical systems. These findings offer important design principles for future aqueous aluminum batteries and mechanically rechargeable aluminum – air energy-storage technologies.

Rubidium Superoxide as a Stable Discharge Product for Metal–O₂ Batteries

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Rechargeable metal–O₂ batteries have attracted considerable attention because oxygen can serve as the positive-electrode active material, offering exceptionally high theoretical energy densities. Their practical development, however, remains limited by the insufficient reversibility of oxygen reduction and evolution reactions. In nonaqueous electrolytes containing alkali-metal ions, the one-electron reduction of oxygen initially produces alkali-metal superoxide (MO₂), whose subsequent behavior depends strongly on its chemical stability. LiO₂ is unstable because the small Li⁺ ion is poorly matched with the much larger superoxide ion, O₂^{•−}. It therefore exists mainly as a transient intermediate and is converted into Li₂O₂ through disproportionation and/or further electrochemical reduction. The resulting peroxide is difficult to reoxidize and causes a large charging overpotential. NaO₂ and KO₂ can be isolated more readily and reoxidized at substantially lower overpotentials.^[1–2] Nevertheless, these superoxides still react gradually with trace H₂O and CO₂, as well as electrolyte components, producing side products that reduce the reversibility of the oxygen electrode. Enhancing the intrinsic stability of MO₂ is therefore an important approach to improving rechargeable metal–O₂ batteries. In this study, oxygen-electrode chemistry was extended to electrolytes containing Rb⁺ and Cs⁺, whose ionic radii are larger than that of K⁺, and the formation and stability of MO₂ were systematically compared across Na, K, Rb, and Cs systems.^[3] RbO₂ and CsO₂ were identified as discharge products and were reoxidized during charging. Among the investigated systems, the Rb-based electrolyte provided the highest reversibility and the most stable cycling behavior, whereas CsO₂ showed evidence of gradual side reactions. Density-functional-theory calculations further indicated that conversion of RbO₂ into bicarbonate is less favorable than the corresponding reactions of the Na, K, and Cs compounds. These results reveal that superoxide stability does not simply increase with alkali-metal ionic radius. Instead, it is maximized when the size of the alkali-metal cation is favorably matched with that of O₂^{•−}, with Rb⁺ providing an optimum balance. Ion-size matching between the cation and superoxide anion thus offers a useful design principle for controlling discharge-product stability and improving the reversibility of metal–O₂ batteries.

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A Gesture Recognition Smart Glove Using Fabric-Based Conductive Carbon Spheres: Self-Powered Zinc-Air Battery System and Its Sensing Performance

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Flexible gesture recognition technology holds broad application potential in human-computer interaction, virtual reality, and smart healthcare. However, existing gesture recognition systems still face numerous challenges in practical applications, including insufficient sensor sensitivity, significant response delays, limited energy supply, and poor adaptability in complex environments, which severely restrict practical applications. In this presentation, micro/nano conductive carbon spheres and hollow carbon spheres were synthesized using a gradient high-temperature annealing method. These were combined with a PVA/fabric substrate to construct micro/nano conductive carbon sphere/PVA/fabric electrodes. Dendritic microstructures and PANI/PVA cylindrical arrays were grown on the fabric electrode surface to fabricate a highly sensitive interlocking pressure sensor. To further enhance sensor performance, the Fowler-Nordheim (F-N) quantum tunneling effect was introduced, leading to the fabrication of an ultra-sensitive multifunctional sensor. Using micron-sized hollow carbon spheres as the framework, $\text{Co}_3\text{O}_4@\text{HCS}$ with needle-like and cubic morphologies was synthesized via hydrothermal growth and gradient high-temperature annealing. The oxygen evolution reaction (OER) and oxygen reduction reaction (ORR) performances were further improved by incorporating $\text{Fe}_2\text{Mo}_3\text{O}_8/\text{MoS}_2$, resulting in the successful assembly of high-performance zinc-air batteries. The fabric sensors and zinc-air batteries were synergistically integrated into a data glove, enabling the full visualization system integration for gesture recognition and offering a new technical pathway and system-level solution for next-generation wearable human-computer interaction devices.

High-Energy Lithium Batteries: Synergistic Materials Design and Closed-Loop Recycling

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This work addresses the growing demand for high-energy-density lithium batteries through integrated advances in materials engineering, interfacial optimization, and closed-loop recycling. Precision carbon coating and heteroatom doping were employed to enhance the capacity and rate capability of $\text{LiMn}_{0.6}\text{Fe}_{0.5}\text{PO}_4$ (LMFP) cathodes, which, when paired with Li_5FeO_4 lithium supplementation agents and high-capacity Si/C anodes, delivered substantially improved full-cell energy density and cycling stability. Interfacial stability under high voltage was achieved via a ternary functional additive strategy that promotes the formation of inorganic-rich CEI/SEI layers, effectively suppressing cathode degradation and dendrite growth. On the sustainability front, a wet chemical directional etching method was developed to directly regenerate spent cathode black mass into structurally intact single-crystal cathodes with simultaneous surface coating and bulk ion doping, yielding performance matching or exceeding commercial counterparts. This study provides a comprehensive, upcycling-oriented pathway for high-energy lithium batteries, bridging materials innovation with end-of-life recyclability.

Boosting the performance of organic cathode materials based on redox-active covalent organic frameworks for metal-ion batteries

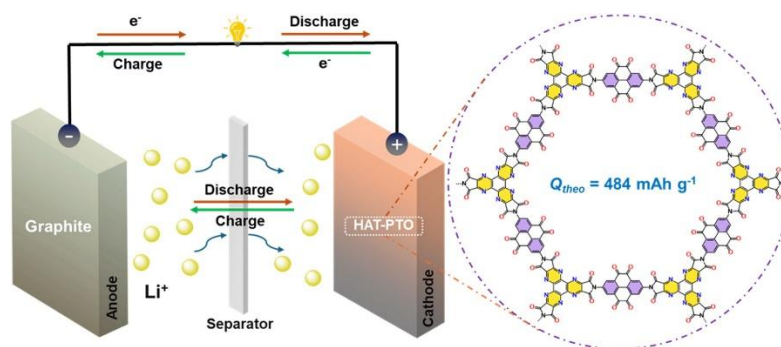
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Electroactive organic materials have received much attention as alternative electrodes for metal-ion batteries due to their high theoretical capacity, resource availability, and environmental friendliness. Redox-active covalent organic frameworks (COFs) have recently emerged as promising electrodes due to their tunable electrochemical properties, insolubility in electrolyte, tunable porosity, and structural versatility.^[1] In this presentation, I will show some recent strategies in terms of molecular design to increase the specific capacity or voltage of some COF-based electrodes. In particular, some examples of electrode materials based on n-type COFs used as high-capacity organic cathodes in Li and beyond-Li (Na, Mg, Al) batteries will be presented.^[2,3,4,5] On the other hand, the electrochemical performance of high voltage organic cathodes based on tetrathiafulvalene-based COFs will be discussed.^[6] Finally, I will show some approaches to increasing the electrical conductivity of COFs, one of the main limitations of these materials for use in energy storage.^[7] In particular, some examples of semiconductor COFs based on the incorporation of neutral organic radicals will be presented.^[8]



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**Brief Ball Milling Coupled with Low-Dose Molten Salt Recrystallization:
Synchronous Particle Homogenization and Intrinsic Al Impurity In-situ Doping
toward High-Voltage Stable Recycled LiCoO₂**

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The rapid iteration of consumer electronics generates massive spent LiCoO₂ cathodes, causing strategic Li/Co resource shortages and severe heavy metal solid waste pollution. Current direct regeneration methods require additional purification to remove intrinsic Al impurities from cathode black mass, which increases manufacturing costs and induces irreversible loss of valuable metallic components. Herein, we propose an integrated regeneration strategy via short-time ball milling coupled with low-dose molten salt, which synchronously achieves grain homogenization and in-situ functional doping of intrinsic Al impurities in spent cathode materials. Short-time ball milling effectively refines particle grains and shortens Li⁺ diffusion pathways. Different from conventional excessive molten salt systems that bring severe surface lattice degradation during repeated water washing, the optimized low-dose molten salt constructs a uniform high-temperature liquid environment to promote homogeneous Al lattice incorporation and suppress surface structural defects. Combined with multi-scale characterizations including in-situ XRD, STEM-EELS and XPS, the lattice stabilization mechanism induced by Al doping is systematically elucidated. Electrochemical evaluations under 4.5 V high-voltage conditions demonstrate that the regenerated LiCoO₂ exhibits superior electrochemical performance to commercial counterparts. It delivers a high discharge capacity of 172.4 mAh g⁻¹ at 5C and maintains a capacity retention of 97.1% after 100 cycles at 0.33C. This innovative upcycling strategy converts detrimental Al impurities into effective lattice stabilizers, breaking the conventional “impurity elimination prior to material repair” paradigm. It provides a low-cost, scalable pathway for the high-value closed-loop regeneration of industrial spent LiCoO₂ and offers universal guidance for the upcycling of various layered cathode materials.

Toward Co-Free Single-Crystal Ni-Rich Cathodes: Challenges and Progress

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Ni-rich layered oxides are promising cathode materials for high-energy-density lithium-ion batteries, yet their practical development remains restricted by structural instability, interfacial degradation, and progressive performance decay. Reducing or eliminating Co offers advantages in terms of cost, resource availability, and sustainability. However, Co removal also alters phase formation, Li-ion transport, redox kinetics, and structural reversibility, creating challenges for the design of stable Ni-rich cathodes. These effects are particularly relevant to single-crystal materials, in which the mitigation of intergranular cracking does not prevent degradation within the bulk structure or at the electrode–electrolyte interface. This presentation summarizes our recent progress in the design and development of single-crystal Ni-rich cathodes based on NM, NMA, and NCMA compositions. The relationships among composition, synthesis conditions, crystal growth, particle morphology, cation mixing, and electrochemical behavior are examined to clarify the factors governing the formation and performance of these materials. Particular attention is directed toward understanding how Co removal modifies their physicochemical and electrochemical characteristics such as reaction reversibility and suppressing structural deterioration and also how Mn and Al influence the stability of the Co-free compositions. Among the Co-free compositions, Al-containing NMA exhibits greater structural and electrochemical stability than binary NM, underscoring the importance of compositional regulation beyond simply eliminating cobalt. Capacity, rate capability, cycling stability, reaction kinetics, and structural reversibility are studied alongside post-cycling structural, morphological, and interfacial analyses. These provide insight into degradation pathways involving surface reconstruction, particle damage, impedance growth, and irreversible structural evolution. The remaining challenges associated with controlling single-crystal formation, preserving structural integrity, and mitigating surface and interfacial degradation are also discussed. This work highlights both the potential and the limitations of removing Co from single-crystal Ni-rich layered cathodes and identifies compositional and synthetic considerations for the continued development of stable, practically relevant Co-free cathode materials.

Engineering fibril networks and microstructure evolution in semi-dry LFP electrodes via extrusion-assisted processing

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Solvent-free (dry) electrode manufacturing is a promising route to reduce the cost, energy consumption, and environmental impact of lithium-ion battery production [1]; however, achieving mechanically robust and electrochemically performant films at high solid contents remains challenging [2]. In this study, we developed a semi-dry processing method for Lithium Iron Phosphate electrodes using a twin-screw extrusion-assisted fibrillation approach with Polytetrafluoroethylene and Ketjen black conductive additives at high solid loading (~85 wt.%, targeting >90 wt.% solid content in the final formulation).

A sequential mixing strategy involving dry premixing, semi-dry extrusion, temperature-controlled multipass processing, and roll-mill film formation was developed to promote PTFE fibrillation and improve particle network homogeneity. The influence of extrusion temperature and processing passes on microstructural evolution, mechanical integrity, and electrochemical behavior was systematically analyzed. SEM observations revealed the progressive development of the fibril network and improved particle interconnections with increasing extrusion temperature and processing intensity. Analysis of the mechanical properties demonstrated notable changes in the tensile behavior and storage modulus, indicating a strong dependence of the film cohesion on the fibrillation state and processing history.

The fabricated freestanding electrodes exhibited stable electrochemical activity in a half-cell configuration, delivering reversible capacities exceeding 140 mAh g⁻¹ at 0.1 C with well-defined redox behavior. Preliminary cycling results demonstrated improved electrochemical stability under optimized extrusion conditions. Ongoing work focuses on correlating porosity, tortuosity, and ionic transport characteristics with processing-induced microstructural changes using impedance analysis and advanced electrode characterization. Single-layer pouch-cell fabrication and further mechanical reliability studies are in progress to evaluate their practical applicability.

This study provides insights into the relationship between semi-dry processing conditions, PTFE fibrillation, electrode architecture, and electrochemical performance, contributing to scalable solvent-reduced manufacturing strategies for next-generation lithium-ion batteries.

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Development of a Continuous High-Speed Conductive Carbon Coating Process for NCM Cathode Active Materials with a Continuous Pre-Mixing

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The surface distribution of conductive carbon in cathode active materials is a critical factor in dry electrode manufacturing, because it directly affects the electronic conductivity, interparticle contact, and fibrillation behavior of polytetrafluoroethylene (PTFE) binders. Conductive carbon attached to the active material surface can improve electronic conduction pathway and may also serve as an anchoring site that promotes PTFE fibrillation during the dry kneading process. Therefore, the development of a scalable and continuous carbon-coating process is an important step toward practical dry-electrode manufacturing.

Conventional conductive carbon coating and composite-powder preparation processes are commonly performed using batch-type mixers or high-energy mixing devices. Although these methods can provide effective carbon distribution at a relatively small scale, their limited process continuity and scalability remain challenges for large-scale mass production. In this study, a continuous high-speed coating process was investigated using a lab-scale twin-screw extruder. Polycrystalline NCM811, C-65 conductive carbon, and PTFE were used as the cathode active material, conductive additive, and binder, respectively. NCM811 and C-65 were lightly premixed and continuously processed under different twin-screw operating conditions.

The degree of conductive carbon attachment to the NCM811 surface was evaluated using scanning electron microscopy (SEM), particularly after the subsequent kneading process with PTFE. The electrochemical performance was examined to assess the electronic conduction network of the resulting dry electrodes. The electrodes prepared through the continuous twin-screw coating route maintained a discharge capacity of over 180 mAh g⁻¹ at 0.1C. SEM observations further confirmed that a significant amount of conductive carbon remained attached to the NCM811 surface even after kneading, indicating that the twin-screw process induced meaningful carbon attachment, rather than simple powder mixing.

These findings demonstrate the feasibility of using a twin-screw extruder as a continuous and high-throughput platform for the conductive carbon coating of cathode active materials. The proposed process provides a promising basis for scaling up the production of coated active materials and contributes to the development of continuous, solvent-free, and mass-producible dry technologies.

Unravelling the Role of Doping in Prussian Blue Frameworks for Alkali-based Batteries

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As global energy production and storage demands continue to rise, the development of sustainable alternatives to lithium-ion batteries (LIBs) has become urgent. Sodium-ion batteries (SIBs) have emerged as a promising low-cost and sustainable technology due to the natural abundance and wide availability of sodium. [1] However, its larger ionic radius hampers the direct transfer of LIB materials to SIBs and poses major challenges in identifying cathodes capable of tolerate the significant volumetric changes occurring during sodiation without compromising structural stability. Among all, Prussian blue analogues (PBAs) have emerged as a promising class of compounds thanks to their water-based, easily scalable synthetic procedure, low cost, and reduced reliance on critical raw materials. Nevertheless, PBAs often suffer from vacancies and structural defects associated with the presence of interstitial water within the framework, which can negatively affect cycling stability and long-term performance. [2]

In this study, a series of doped PBAs were synthesized through the incorporation of divalent and trivalent ions as dopants, and comprehensively characterized. The best candidate has resulted in improved performance, delivering a high specific capacity of 140 mAh g⁻¹ at C/8, outperforming the pristine material (120 mAh g⁻¹), while maintaining remarkable stability over 3300 cycles at 2C with minimal capacity fading. Operando XRD, ex situ XPS, SEM, TEM, CV, and PEIS analyses confirmed enhanced structural integrity during cycling and suggested a greater contribution from low-spin iron centres, which is proposed to play a key role in the improved electrochemical performance. Furthermore, the increased Na⁺ diffusion coefficient supports the beneficial effect of doping in promoting fast and reversible sodium insertion/extraction, enhancing the overall performance of PBA cathodes.

Atomistic Investigation of Fe-Substituted Mixed Polyanionic Cathodes for Sodium-Ion Batteries

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Mixed-polyanionic phosphates are promising cathode materials for sodium-ion batteries owing to their high operating voltages, thermal stability, and robust crystal frameworks. However, $\text{Na}_4\text{Mn}_3(\text{PO}_4)_2(\text{P}_2\text{O}_7)$ suffers from Mn^{3+} -induced Jahn-Teller distortions during desodiation, leading to structural instability and capacity degradation. Understanding the effect of Fe substitution in $\text{Na}_4\text{Mn}_{3-x}\text{Fe}_x(\text{PO}_4)_2(\text{P}_2\text{O}_7)$ ($0 \leq x \leq 3$) is therefore essential for designing high-performance mixed-polyanionic cathodes. In this work, we develop a computationally efficient framework for investigating this important class of materials under experimentally relevant conditions. By combining a newly developed classical interatomic potential with density functional theory (DFT), enables large-scale atomistic simulations at realistic time and length scales that are beyond the reach of first-principles methods alone. This framework provides a comprehensive understanding of the structural, electronic, and ionic transport properties across the full compositional range, enabling systematic investigation of sodium extraction/insertion, phase evolution, and whether the charge-discharge process proceeds through a single-phase or two-phase reaction mechanism. Our results show that progressive Fe substitution suppresses Mn^{3+} -induced Jahn-Teller distortions by modifying the local crystal-field environment and transition-metal-oxygen bonding, reducing the desodiation-induced volume change from $\sim 6.2\%$ for the Mn-rich composition to $\sim 3.05\%$ for the Fe-rich end member. Electronic structure analysis reveals that the $\text{Fe}^{2+}/\text{Fe}^{3+}$ redox couple enhances charge delocalization, providing an atomistic explanation for the improved structural stability. While increasing Fe content lowers the average intercalation voltage from 3.7 to 3.2 V, the combined atomistic analysis identifies an optimal Fe concentration that balances electrochemical performance, structural integrity, and reversible sodium storage. Together, these results demonstrate the potential of multiscale atomistic modeling to uncover composition-structure-property relationships and guide the discovery of advanced sodium-ion battery cathodes.

Micron-scale Fe₂P Modified with an N-doped Carbon Layer for Efficient Polysulfide Regulation and Sulfur Redox Conversion

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Lithium–sulfur batteries offer high theoretical energy density but still suffer from polysulfide crossover and kinetically sluggish sulfur conversion reactions. In this work, a functional separator incorporating micron-scale Fe₂P modified with a polydopamine-derived N-doped carbon layer (Fe₂P@NC) is introduced to address these limitations. Fe₂P serves as a polar catalytic component that chemically interacts with lithium polysulfides, whereas the attached N-doped carbon layer improves electron transport and stabilizes the particle surface. The resulting interlayer combines polysulfide confinement with catalytic promotion of sulfur redox reactions. Li₂S₆ diffusion and adsorption experiments demonstrate that the Fe₂P@NC separator effectively restricts polysulfide migration. Electrochemical measurements using symmetric cells further reveal enhanced polysulfide conversion activity. In full cells, the Fe₂P@NC separator lowers electrochemical polarization, as indicated by a decrease in the anodic–cathodic peak separation from 0.327 V for the conventional separator to 0.256 V. The improved reaction kinetics enable a discharge capacity of 639 mAh g^{−1} at 5 C. In addition, the cell retains 72.4 % of its initial capacity after 100 cycles at 1 C. These results highlight N-doped carbon surface modification of micron-scale Fe₂P as a practical approach for developing conductive and catalytically active separators for high-rate, long-term lithium–sulfur batteries.

A Scalable Thermal Prelithiation Strategy Using Air-Stable Lithium

Precursors for High-Performance Silicon Anodes

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Silicon-based anodes are considered highly attractive for next-generation lithium-ion battery (LIB) technologies owing to their exceptionally high theoretical capacity, low cost, and natural abundance. Nevertheless, their commercial viability is limited by poor initial Coulombic efficiency (ICE) and continuous degradation of the electrode/electrolyte interface. In this study, a simple and scalable solid-state thermal prelithiation strategy employing low-cost and air-stable lithium precursors (LiOH and Li₂CO₃) is proposed to improve the electrochemical performance of the Si anodes. The effects of thermal treatment temperature (800–1000 °C) and lithium precursor chemistry on phase evolution, lithiation behavior, and electrochemical properties are systematically investigated. The optimized calcination temperature of 900 °C creates a balanced lithium silicate interphase with a high degree of prelithiation while suppressing undesirable SiO₂ formation. Furthermore, precursor-dependent interfacial chemistries are observed. LiOH promotes the formation of a Li₄SiO₄-rich interphase with enhanced Li⁺ transport, resulting in superior rate capability, whereas Li₂CO₃ generates a more stable interphase that improves cycling durability. The optimized Si–LiOH anode demonstrates an ICE of approximately 90%, a reversible capacity of 1140 mAh g^{−1} at 5 A g^{−1}, and stable long-term cycling. Benefitting from its powder-level processing and compatibility with conventional LIB manufacturing, this air-stable thermal prelithiation approach offers a practical and cost-effective pathway towards scalable production of high-performance silicon anodes for advanced LIBs.

Keywords: Silicon anodes; Thermal prelithiation; Lithium silicate; Air-stable lithium precursors; Lithium-ion batteries

Novel Additive for Addressing Interfacial Instability in PVDF-HFP/Pyr₁₄FSI Systems for Lithium Metal Batteries at Room Temperature

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Solid-state secondary batteries are emerging as the next-generation electrochemical energy storage systems, offering high performance for electric vehicles and smart grid applications. To facilitate a sustainable energy transition, the development of safe and reliable solid polymer electrolytes (SPEs) is crucial. These electrolytes not only enhance energy density and safety but also enable the use of lithium metal anodes, thereby advancing more efficient and durable battery technologies [1,2]. The SOLVE European project aims to address key challenges associated with solid polymer electrolytes (SPEs) for next-generation solid-state secondary batteries. The project focuses on the development of mechanically robust macromolecular networks incorporating specific additives or fillers to achieve high ionic conductivity and improved electrochemical stability.

Self-standing hybrid solid polymer electrolytes (HSPEs) are developed herein via a fully solvent-free and energy-efficient process, offering a scalable and industry-compatible route for next-generation solid-state batteries. The proposed HSPEs are based on a poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP)/Pyr₁₄FSI ionic liquid system, incorporating a small amount of divinyl sulfone (DVS) as a functional additive to tailor interfacial and mechanical properties.

This design achieves a balanced combination of high ionic conductivity, robust mechanical integrity, and enhanced thermal stability. The ionic liquid phase ensures efficient ion transport and contributes to maintaining conductivity over a wide temperature range, while the incorporation of low DVS content (up to 5 wt%) effectively stabilizes the solid-electrolyte interphase (SEI) and suppresses common degradation pathways typically observed in PVDF-HFP/Pyr₁₄FSI-based electrolytes. In addition, the presence of DVS appears to improve interfacial compatibility with lithium metal, promoting more uniform plating/stripping behaviour [3,4].

Overall, these results highlight a simple but effective interfacial engineering strategy to enhance the durability and electrochemical stability of ionic-liquid-based polymer electrolytes, paving the way toward safe, high-voltage, and high-performance solid-state lithium metal battery technologies.

Boosting Electrochemical Performance of $\text{Ti}_3\text{C}_2\text{T}_x@\text{Ni}$ MXene-Based Composite Electrodes

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The $\text{Ti}_3\text{C}_2\text{T}_x@\text{Ni}$ composite was prepared using pressureless sintering and in-situ growth techniques, exhibiting superior mechanical structure and electrochemical performance. The in-situ grown Ni nanoparticles serve as interlayer pillars, effectively expanding MXene sheets, increasing interlayer spacing, and inhibiting sheet stacking during cycling, thereby significantly enhancing electrolyte ion diffusion rates. The high conductivity of Ni forms a continuous conductive network, reducing electrode series resistance and interfacial charge transfer resistance while accelerating electrochemical reaction kinetics. Additionally, the introduction of Ni nanoparticles provides abundant pseudocapacitive redox-active sites, enabling synergistic energy storage between the double-layer and pseudocapacitance, resulting in a 29% increase in specific capacitance at 1 A g^{-1} . Benefiting from the strong Ti-O-Ni interfacial chemical bonding structure, the composite maintains a remarkable capacity retention of 97.1% after 8,000 long cycles, far surpassing pure MXene. When used as the cathode in an asymmetric supercapacitor, the device achieves a high energy density of 7.2 Wh kg^{-1} and excellent stability over 10,000 cycles

Keywords: MXene, Electrode, $\text{Ti}_3\text{C}_2\text{T}_x@\text{Ni}$, Hydrothermal method, Supercapacitor

Rational Design of $\text{MnFe}_2\text{O}_4@\text{Ti}_3\text{C}_2\text{T}_x$ MXene

Heterostructures from Bimetallic MOF for High-Capacity Li/Na-ion Batteries

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The development of high-performance electrode materials is the key to future alkali-ion batteries. Herein, a MOF-derived $\text{MnFe}_2\text{O}_4@\text{Ti}_3\text{C}_2\text{T}_x$ MXene heterostructure is reported as a high-performance anode for Na-ion and Li-ion batteries. The strong interfacial interaction between MnFe_2O_4 spindle-shaped nanoparticles and conductive $\text{Ti}_3\text{C}_2\text{T}_x$ nanosheets enhances the electron/ion mobility and structural integrity. The optimized 1:1 ratio of $\text{MnFe}_2\text{O}_4@\text{Ti}_3\text{C}_2\text{T}_x$ composite demonstrates a reversible capacity of 975 mAh g^{-1} at 0.1 A g^{-1} and 290 mAh g^{-1} at 5 A g^{-1} when used in sodium-ion batteries. In lithium-ion batteries, it provides a capacity of 1190 mAh g^{-1} at a current of 0.1 A g^{-1} and maintains 270 mAh g^{-1} at 2 A g^{-1} . In-situ XRD and Raman results confirm the structural stability of the heterostructure after cycling. These results demonstrate a scalable route for the preparation of MOF-derived MXene heterostructures with excellent electrochemical performance for advanced Na-ion and Li-ion energy storage.

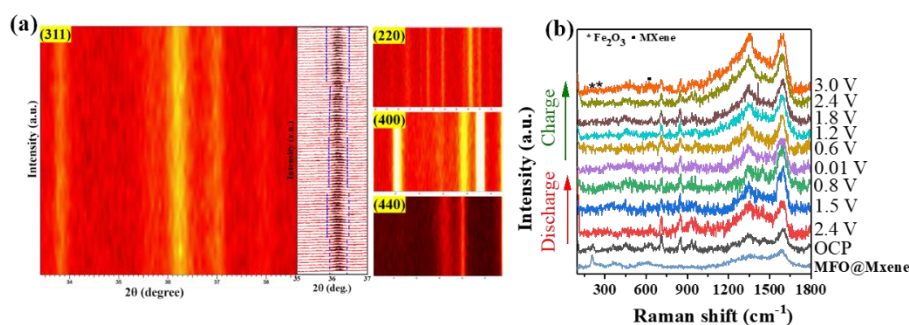


Fig.1 (a) *In-situ* XRD and (b) *In-situ* RAMAN of MFO@MXene for Na-ion battery.

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Additive-Driven Regulating Solvation Structure Toward Stable High-Voltage Lithium Metal Batteries

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Raising the upper cutoff voltage is an effective route to high-energy lithium-metal batteries (LMBs), but severe interfacial instability at Ni-rich cathodes and lithium-metal anodes limits their operation. Conventional additives such as vinylene carbonate (VC), fluoroethylene carbonate (FEC), and lithium bis(oxalato)borate (LiBOB) are usually regarded as sacrificial film-forming agents, while their role in regulating bulk Li^+ solvation remains underexplored. Herein, we develop a ternary-additive carbonate electrolyte and reveal a "solvation reconstruction-interphase coupling" mechanism, supported by spectroscopy, simulations, and interphase analyses. VC/FEC/LiBOB synergistically reshapes the Li^+ primary solvation sheath and tunes theSSIP/CIP/AGG distribution, shifting the electrolyte from a solvent-dominated structure toward an anion-/additive-involved coordination environment without compromising bulk ion transport. Meanwhile, their preferential decomposition constructs inorganic-rich CEI/SEI layers with enhanced mechanical robustness and reduced interfacial resistance, thereby suppressing electrolyte oxidation, cathode degradation, and Li dendrite growth. As a result, $\text{Li}||\text{NCM92}$ cells deliver $>185 \text{ mAh g}^{-1}$ after 50 cycles at 3.0–4.7 V and 150 mAh g^{-1} at 10 C, $\text{Li}||\text{Li}$ cells operate stably for $>1300 \text{ h}$, and $\text{Gr}||\text{NCM811}$ pouch cells retain 90% capacity after 100 cycles. This work redefines conventional film-forming additives as dual-function solvation/interphase regulators for high-voltage LMBs.

Structural and Interfacial Engineering of Ceramic-Coated Separators for Stable Lithium Metal Batteries

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Lithium metal batteries offer high energy density but suffer from nonuniform Li^+ transport, dendritic lithium growth, unstable interfacial reactions, and internal short circuits. Here, we present two complementary ceramic-coated separator strategies that regulate lithium deposition through structural and interfacial engineering.

A dry-biaxially stretched polypropylene separator with a highly porous, low-tortuosity pore network enabled rapid Li^+ transport but exhibited limited resistance to dendrite penetration. A nanometer-scale, binder-free Al_2O_3 coating was therefore introduced by radio-frequency sputtering. The ultrathin coating preserved the intrinsic pore structure while improving electrolyte wettability, Li^+ flux uniformity, and short-circuit resistance, resulting in enhanced high-rate capability and cycling stability.

In a complementary approach, an AlN-based ceramic-coated separator was designed to regulate Li-metal interfacial chemistry and thermal behavior. The AlN coating induced the formation of a Li_3N -rich ion-conductive interphase and a lithiophilic Li–Al alloy, reducing the nucleation barrier and promoting uniform lithium deposition. Its high thermal conductivity further contributed to the dissipation of localized heat.

These studies demonstrate that ceramic-coated separators can function as active interfaces rather than passive mechanical barriers. Rational control of separator pore structure, coating dimensions, and ceramic chemistry enables the concurrent regulation of Li^+ transport, interphase formation, lithium nucleation, and thermal response, providing a practical design strategy for stable and safe lithium metal batteries.

Decoding Electrode Microstructure-Performance Relationships through FIB-SEM-Based 3D Model

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Electrode microstructure critically governs electrochemical performance by determining ionic and electronic transport, reaction heterogeneity, and mechanical stability. However, conventional characterization techniques and low-dimensional models provide limited access to the complex spatial relationships among active materials, pore networks, and carbon-binder domains, making it difficult to establish a correlation between electrode structure and performance. Here, we present a high-resolution FIB-SEM tomography-based framework for constructing microstructure-resolved electrode models. Tomographic images are processed and segmented to reconstruct the electrode microstructure in 3D. Following validation against experimentally measured structural, electrochemical, and mechanical properties, the reconstructed models serve as digital twins for quantifying veiled information, including interfacial contact, pore connectivity, electronic conduction pathways, concentration and overpotential distributions, and stress evolution during electrode operation. This framework enables the diagnosis of transport limitations, reaction heterogeneity, and degradation mechanisms, as well as the identification of electrode design windows that balance electrochemical performance and mechanical stability. It thereby establishes a pathway from high-resolution 3D imaging to physics-based analysis and the predictive design of high-performance, high-energy-density electrodes.

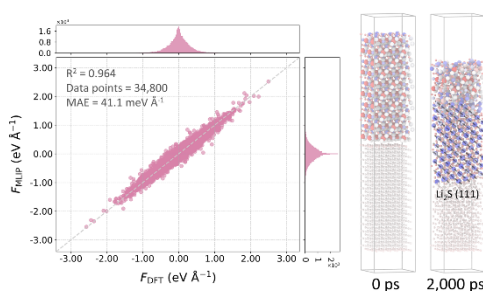
Unveiling Enhanced Li-Ion Mobility in Amorphous SEI Layers: A Machine-Learning-Accelerated Molecular Dynamics Study of the Solid Electrolyte/Li-Metal Interface

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Due to the dramatically increasing energy demand in electric vehicle and data center applications, the development of next-generation energy storage devices with higher energy densities is essential. In particular, Li-metal batteries utilizing solid electrolytes exhibit extremely high volumetric energy density and enhanced safety. The risks of electrolyte leakage, Li dendrite growth, and subsequent short circuits are significantly reduced by the high mechanical modulus and non-flammable nature of solid electrolytes. Although replacing conventional liquid-based electrolytes addresses these safety issues and highlights the promising applicability of all-solid-state Li-metal batteries (ASSLMBs), several inherent drawbacks of solid electrolytes still limit their commercialization. For instance, the sulfide-based argyrodite-type $\text{Li}_6\text{PS}_5\text{Cl}$ (LPSC) suffers from H_2S formation, poor interfacial contact, and slower Li diffusion compared to liquid electrolytes. Cation doping strategies have been employed to mitigate H_2S formation and improve the cycling stability of ASSLMBs. In this work, we examined the interfacial phenomena between the Li-metal anode and Si-doped LPSC through long-term MD simulations based on a fine-tuned machine-learning interatomic potential (MLIP). The formed solid electrolyte interphase (SEI) is highly amorphous, and the formation of a Li-Si alloy is observed. In contrast, the pristine LPSC features a crystalline Li_2S SEI with partial S replacement by Cl and P. We further calculated the Li-ion mobility; the pristine LPSC shows sluggish Li-ion diffusion due to the crystallized SEI, whereas the amorphous SEI in the Si-doped LPSC system exhibits improved Li-ion mobility. This work reveals the effect of cation doping on SEI formation and Li-ion mobility within the SEI layer, enhancing our atomistic understanding of the complex solid electrolyte/Li-anode interface and providing guidance for future materials design.



Addressing Electro-Chemo-Mechanical Coupling Failure at the Sodium|Na₃PS₄ Interface towards Durable All-Solid-State Sodium Metal Batteries

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All-solid-state sodium batteries utilizing sulfide electrolytes and sodium (Na) anodes are promising for energy storage due to their safety and cost-effectiveness. However, their practical implementation is significantly hindered by the interfacial degradation at Na₃PS₄|Na interface, while the resolution strategies remain scarce due to the unclear underlying mechanisms. Herein, operando synchrotron X-ray computed tomography is utilized to reveal an electro-chemo-mechanical coupling failure mechanism. Our results demonstrate the formation of a 65-μm-thick side-reaction layer at the interface within two hours of contact. This electron-conducting but ion-blocking layer causes non-uniform Na electrodeposition, triggering continuous mechanical cracking inward Na₃PS₄ that leads to short-circuit. To address this issue, a thermodynamically stable thin Na₃Sb-NaF functional layer is engineered on Na to protect Na₃PS₄ from being chemically reduced. Its high Na-ion diffusivity ensures uniform ion transport for homogeneous Na electrodeposition, enabling mechanically integrated interface. As a result, symmetric cells sustain over 1000 hours of cycling at 0.1 mA cm⁻² (0.1 mAh cm⁻²), while full cells using NaNi_{1/3}Fe_{1/3}Mn_{1/3}O₂ cathodes achieve 88.3% capacity retention after 300 cycles at 0.2 C, room temperature. Crucially, anode-free all-solid-state batteries exhibit stable cycling for over 60 cycles. This work advances the development of high-energy, durable all-solid-state sodium metal batteries.¹

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Ionic Microenvironment Regulator Enables Enhanced Anion Trapping in Cationic Poly(ionic liquid) Solid Electrolytes for Low-Temperature Lithium Metal

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Despite the considerable promise of solid polymer electrolytes (SPEs) for enabling high-energy-density and intrinsically safe lithium metal batteries, their practical deployment in cold environments remains hindered by sluggish Li^+ transport at sub-zero temperatures. Herein, we incorporate methyl trifluoromethanesulfonate (TMS) into a cationic poly(ionic liquid) solid electrolyte to promote Li^+ transport under extreme low-temperature conditions. Notably, TMS not only enhances lithium salt dissociation but also induces an anion-trapping effect by modulating the local charge distribution of the polycationic backbone, thereby facilitating uniform lithium deposition. As a result, the optimized cationic poly(ionic liquid) solid electrolyte (PIL-TMS₁) enables stable cryogenic operation of high-voltage $\text{Li}||\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ and $\text{Li}||\text{LiFePO}_4$ batteries, delivering capacity retentions of 89.8% after 500 cycles and 80.1% after 800 cycles at $-15\text{ }^\circ\text{C}$, respectively. This work presents an effective strategy for designing low-temperature solid-state lithium metal batteries through additive-regulated anion trapping and accelerated Li^+ transport.

Engineering Layered Zn(002) via Oscillation-Regulated Electrodeposition on Reconstructed Zn Substrates for Stable Aqueous Zinc-Ion Batteries

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Aqueous Zn-ion batteries are promising for safe and low-cost energy storage, but their practical deployment remains limited by poor Zn anode reversibility arising from dendritic growth, parasitic reactions, and interfacial instability. In this work, we present a strategy that combines crystallographic reconstruction with oscillation-regulated electrodeposition to stabilize Zn metal anodes. Mechanical polishing reconstructs commercial Zn foils into crystallographically textured Zn substrates, providing a favorable platform for subsequent Zn growth. On this reconstructed surface, oscillation-assisted electrodeposition regulates interfacial processes during plating and directs Zn growth into a layered hexagonal architecture. This regulated growth mode suppresses hydrogen evolution, corrosion, and polarization accumulation, leading to enhanced Zn plating/stripping reversibility and stable symmetric cell operation for over 1000 h at 50 mA cm⁻² with 10 mAh cm⁻². When paired with a Mn(II)-expanded hydrated vanadium oxide cathode, the resulting full cells deliver improved rate capability and 88% capacity retention after 1000 cycles at 4 A g⁻¹. This work highlights how crystallographic reconstruction and regulated interfacial growth can be coupled to improve Zn anode stability and provides a simple route toward durable aqueous Zn-ion batteries.

Keywords: Zn anode, Electrochemical oscillation, Zinc-ion battery and Dendrite-free

High-Energy-Density Solid-State Lithium Batteries Enabled by Ultrathin Composite Solid Electrolyte and High-Loading Composite Cathodes

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Solid-state lithium batteries (SSLBs) have attracted significant attention owing to their high energy density and enhanced safety. However, their practical application is hindered by the large thickness and limited ionic conductivity of composite solid electrolytes (CSEs), as well as high electrode/electrolyte interfacial resistance. In this study, an 18 μm -thick CSE was developed using a polyethylene scaffold integrated with garnet-type $\text{Li}_{6.25}\text{La}_3\text{Zr}_2\text{Ga}_{0.25}\text{O}_{12}$ (LLZGO) and an ionic liquid (IL) additive within a poly(vinylidene fluoride-co-hexafluoropropylene)/polypropylene carbonate matrix. The resulting CSE exhibits a high ionic conductivity of $8.6 \times 10^{-4} \text{ S cm}^{-1}$ at 30 $^{\circ}\text{C}$. The incorporation of IL enhances Li^+ transport and effectively reduces the interfacial resistance. To further improve the cell-level energy density, LLZGO and *N*-propyl-*N*-methylpyrrolidinium bis(trifluoromethanesulfonyl)imide (PMP-TFSI) IL are incorporated into a high-loading $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ (NCM-811) cathode. The synergistic effects of LLZGO and PMP-TFSI within the cathode facilitate Li^+ transport and improve electrode performance. A thick cathode with an active material loading of approximately 20 mg cm^{-2} is successfully fabricated, delivering an areal capacity of $\sim 4 \text{ mAh cm}^{-2}$. By combining the scaffold-supported thin CSE with the thick NCM-811||LLZGO||PMP-TFSI composite cathode, the projected energy density of an anode-free pouch cell reaches $\sim 420 \text{ Wh kg}^{-1}$. These results demonstrate a scalable strategy for constructing oxide-based SSLBs with high energy density, enhanced Li^+ transport, and strong potential for practical applications.

Spray-Dried Microsized Porous Carbon Host with C-Doped g-C₃N₄ for High-Performance Sulfur Cathodes

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Lithium–sulfur batteries have attracted considerable attention owing to their high theoretical specific capacity of 1675 mAh g⁻¹ and high energy density. However, their practical application remains challenging because of the low electrical conductivity of sulfur and the dissolution of lithium polysulfides (LiPSs) during cycling. Porous carbon materials have been widely investigated as sulfur hosts due to their high surface area and pore volume. Nevertheless, the low tap density of nanosized carbon materials limits electrode density and consequently hinders the realization of high energy density electrodes.

In this study, a microsized porous carbon host was constructed from nanosized carbon particles through a spray-drying process to improve electrode density while retaining a porous structure for efficient sulfur accommodation. Graphitic carbon nitride (g-C₃N₄), which possesses abundant nitrogen-containing polar sites, was incorporated into the carbon host to enhance chemical interactions with LiPSs and suppress their dissolution. Furthermore, carbon doping of g-C₃N₄ was introduced to improve its electrical conductivity while maintaining the LiPS anchoring capability of the nitrogen-rich sites. This combined strategy integrates the structural advantages of the spray-dried microsized porous carbon host with the chemical affinity and enhanced electronic conductivity of C-doped g-C₃N₄.

Electrochemical analysis demonstrated that the incorporation of C-doped g-C₃N₄ improved the electrochemical performance of the sulfur cathode, particularly under high-rate conditions, by facilitating charge transfer and suppressing LiPS dissolution. The enhanced rate capability indicates that simultaneously regulating LiPS confinement and electronic conductivity is critical for achieving efficient sulfur utilization at increased current densities. These results demonstrate that spray-dried microsized porous carbon hosts incorporating C-doped g-C₃N₄ provide an effective strategy for developing high-density and high-performance lithium–sulfur battery cathodes.

Theoretical and Experimental Investigation of W-Substituted P2-Type $\text{Na}_{0.75}\text{Mn}_{0.60}\text{Ni}_{0.30}\text{Fe}_{0.10}\text{O}_2$ Cathode Materials for Sodium-Ion Batteries

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W-substituted P2-type $\text{Na}_{0.75}\text{Mn}_{0.60}\text{Fe}_{0.30}\text{Ni}_{0.10}\text{O}_2$ cathode materials for sodium-ion batteries were synthesized via a ball-milling method. The crystal structure and surface morphology of the synthesized materials were systematically characterized using X-ray diffraction (XRD) and field-emission scanning electron microscopy (FE-SEM). To gain further insight into the effect of W substitution, first-principles density functional theory (DFT) calculations were performed to investigate the structural evolution of the substituted cathodes, while molecular dynamics (MD) simulations were employed to evaluate Na-ion diffusion behavior and diffusion coefficients. Electrochemical measurements revealed that the pristine $\text{Na}_{0.75}\text{Mn}_{0.60}\text{Fe}_{0.30}\text{Ni}_{0.10}\text{O}_2$ electrode delivered an initial discharge capacity of 91 mAh g^{-1} at a 0.5 C rate, which decreased to 68 mAh g^{-1} after 100 charge-discharge cycles. In contrast, the W-substituted electrode exhibited a significantly higher initial discharge capacity of 123 mAh g^{-1} at the same current rate and maintained a reversible capacity of 77 mAh g^{-1} after 100 cycles. The combined experimental and theoretical results indicate that W substitution enhances the structural integrity and electrochemical stability of the P2 framework by mitigating the Jahn-Teller distortion associated with Mn^{3+} and promoting more favorable Na-ion diffusion. Consequently, W substitution represents an effective strategy for simultaneously improving the capacity, cycling stability, and Na-ion transport kinetics of P2-type $\text{Na}_{0.75}\text{Mn}_{0.60}\text{Fe}_{0.30}\text{Ni}_{0.10}\text{O}_2$ cathodes, highlighting their potential for high-performance sodium-ion battery applications.

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MOF-Derived Co₂P-Embedded CNF Interlayers for High-Performance Lithium–Sulfur Batteries

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The lithium–sulfur (Li–S) battery, a promising next-generation energy storage system, is attracting significant attention due to its potential for delivering high energy density at a relatively low cost. This is largely attributed to sulfur's natural abundance, low price, and high theoretical capacity of 1675 mAh/g. However, key challenges remain, including sulfur's intrinsic low conductivity, substantial volume expansion, and the notorious polysulfide shuttle effect.

To address these issues, many researchers have explored the introduction of interlayers as a viable strategy. Interlayers can mitigate the shuttle effect of polysulfides and improve both electronic and ionic conductivities.

In this study, carbon nanofiber (CNF) was employed as the interlayer material due to its excellent electrical conductivity, and it was fabricated via electrospinning followed by carbonization of polyacrylonitrile (PAN). To further enhance battery performance, cobalt phosphide (Co₂P) and carbon nanotubes (CNTs), derived from metal-organic frameworks (MOFs), were incorporated into the CNF matrix to provide both catalytic functionality and strong physical adsorption. Co₂P was synthesized using ZIF-67 and phytic acid, while thermal treatment of ZIF-67 promoted in-situ CNT growth through catalytic action.

This CNF-based composite interlayer effectively suppresses the polysulfide shuttle and enhances electronic and ionic transport, thereby significantly improving the overall performance of Li–S batteries. The structural and surface properties of the interlayers were characterized using SEM, STEM/EDS, and XRD analyses.

Finally, the fabricated Co₂P–CNT–CNF interlayer was integrated into Li–S cells, where it delivered enhanced electrochemical performance. Diffusion tests confirmed that the shuttle effect was successfully suppressed, validating the effectiveness of Co₂P–CNT–CNF as a functional interlayer for high-performance lithium–sulfur batteries.

Multifunctional Roles of Oxide Additives in Dry Electrodes

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The growing adoption of electric vehicles has driven the demand for high-energy-density lithium-ion batteries. Achieving such high energy densities requires thick electrodes with high active material loading, making dry electrode technology an increasingly attractive manufacturing strategy. Although reducing the PTFE binder content is considered a promising strategy for complying with forthcoming PFAS regulations and increasing the active-material fraction, it inevitably undermines the structural integrity of free-standing dry electrodes, posing a significant challenge for practical manufacturing. Therefore, developing multifunctional additives capable of simultaneously reinforcing electrode integrity and facilitating electrochemical kinetics has become increasingly important. Herein, we systematically investigated the cascading effects of a perovskite oxide additive on (i) the mechanical stability and microstructural evolution of free-standing and laminated dry electrodes, (ii) the buffering of oxygen release from a Li-rich Mn-based cathode, and (iii) the enhancement Li-ion transport across the cathode–electrolyte interface, leading to improved electrochemical performance.

Universal Testing Machine(UTM) and micro-indentation measurements confirmed that the perovskite oxide additive enhanced the tensile strength and hardness of the dry electrodes, whereas ionic resistivity, Li-ion diffusion coefficient, and Laser-Induced Breakdown Spectroscopy(LIBS) analyses provided complementary evidence that the optimized additive content and the associated oxygen-vacancy concentration facilitated Li-ion transport.

Reliable Evaluation of Materials in Na-Ion Batteries Using NASICON Electrode

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Na-ion batteries (SIB) have emerged as promising candidates for next-generation energy storage systems due to their cost-effectiveness and the abundant availability of Na resources.¹ In parallel with the rapid expansion of research into electrode materials for SIB, there has been growing concern about the reliability of the conventional evaluation method using Na metal as a counter electrode. A significant number of studies continue to employ Na metal as a counter electrode in half-cell configurations. However, Na metal is inherently unstable and prone to parasitic reactions with electrolytes, solid-state interphase formation/degradation, and dendrite formation, which can lead to severe irreproducibility and a misleading interpretation of electrochemical performance.

In this work, we critically examine the impact of using Na metal as a counter electrode on the evaluation of electrode and electrolyte materials. By analyzing the effects of Na instability on performance metrics, we provide direct evidence of its limitations in material evaluation. Furthermore, we propose a practical and reproducible framework utilizing NASICON (Na super ionic conductor)-type counter electrodes that eliminate the adverse effects of Na metal, while enabling an accurate and meaningful assessment, thereby directly supporting the construction of a full cell.²

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Correlating Structure and Na-Ion Diffusion in Ti/Mg Co-Doped P2-Type $\text{Na}_{0.67}\text{Mn}_{0.60}\text{Ni}_{0.20}\text{Fe}_{0.20}\text{O}_2$ Cathodes through Experimental and First-Principles Studies

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P2-type $\text{Na}_{0.67}\text{Mn}_{0.60}\text{Ni}_{0.20}\text{Fe}_{0.20}\text{O}_2$ is considered a promising layered oxide cathode for sodium-ion batteries owing to its high theoretical capacity and low cost. However, severe Jahn-Teller distortion associated with Mn^{3+} and sluggish Na^+ diffusion kinetics lead to structural instability and poor cycling performance, limiting its practical application. Herein, Ti/Mg co-doped P2-type $\text{Na}_{0.67}\text{Mn}_{0.60-2x}\text{Ti}_x\text{Mg}_x\text{Ni}_{0.20}\text{Fe}_{0.20}\text{O}_2$ ($x = 0.01, 0.02$, and 0.05) cathode materials were synthesized via a conventional ball-milling method. The crystal structure, morphology, and electrochemical performance of the prepared samples were systematically investigated. Furthermore, first-principles calculations, including density functional theory (DFT), the nudged elastic band (NEB) method, and *ab initio* molecular dynamics (AIMD) simulations, were employed to elucidate the effects of Ti/Mg co-doping on structural stability, Na^+ migration barriers, and diffusion behavior. The incorporation of electrochemically inactive Ti^{4+} and Mg^{2+} effectively suppresses Jahn-Teller distortion, enlarges the interlayer spacing, and facilitates Na^+ transport, thereby enhancing the structural stability of the P2 framework. Consequently, the optimized cathode delivers an initial discharge capacity of 123 mAh g^{-1} at 0.5 C and retains 98 mAh g^{-1} after 100 cycles, demonstrating significantly improved cycling stability and rate capability. These findings indicate that Ti/Mg co-substitution is an effective strategy for simultaneously enhancing the structural integrity and electrochemical performance of P2-type $\text{Na}_{0.67}\text{Mn}_{0.60}\text{Ni}_{0.20}\text{Fe}_{0.20}\text{O}_2$ cathodes and provide valuable insights into the rational design of high-performance sodium-ion batteries.

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Development of an elastic polymer protective layer incorporating crosslinker-functionalized inorganics for Li-metal batteries

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Lithium metal anodes have attracted considerable attention as a promising anode for next-generation high-energy-density rechargeable batteries owing to their ultrahigh theoretical capacity (3860 mAh g⁻¹, approximately ten times that of graphite) and extremely low electrochemical potential (-3.04 V vs. SHE). However, during cycling, non-uniform Li deposition promotes dendritic growth and induces large volume fluctuations, which not only accelerates performance degradation but also raises safety concerns due to internal short circuits. Therefore, it is essential to introduce a surface protective layer that can simultaneously suppress dendrite formation and accommodate volume changes.

Elastic polymer-based protective layers can buffer repeated expansion and contraction however, their limited mechanical strength often fails to effectively prevent dendrite penetration. In this study, we developed an organic-inorganic hybrid protective layer in which silica particles are chemically coupled with polymer chains. The silica particles were functionalized with C=C-bearing organic groups to strengthen interfacial bonding and suppress delamination even under mechanical deformation. For practical implementation, a transfer-coating process was further adopted to minimize direct contact between Li metal and the solvent while ensuring compatibility with large-area manufacturing. As a result, Li metal anodes protected with this layer exhibited lower overpotentials and more stable cycling behavior in Li plating/stripping tests compared with bare Li, confirming effective interfacial stabilization. Moreover, the cycle life was improved by approximately twofold in half-cells employing a high-nickel cathode. Overall, this work presents an effective surface-protection strategy for stabilizing Li metal anodes and enabling high-energy-density rechargeable batteries.

Keyword: Lithium metal anode, elastic hybrid polymer protective layer, crosslinker-modified, transfer printing

A Study on the Dry Fabrication Process of High Energy Density Anodes for LFP Batteries

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The demand for lithium-ion batteries with lower internal resistance, higher energy density, improved mechanical stability, and enhanced manufacturing productivity has accelerated the development of dry electrode fabrication technologies as an alternative to conventional wet coating processes. In conventional wet electrode manufacturing, electrode slurries containing active materials, conductive additives, and binders are coated onto current collectors, followed by solvent evaporation through a drying process. However, this approach requires substantial energy consumption for solvent removal, resulting in high manufacturing costs and reduced productivity. Furthermore, the emission of volatile organic compounds (VOCs) during drying raises environmental concerns, while solvent migration, binder segregation, and crack formation limit the fabrication of thick electrodes with coating thicknesses exceeding approximately 150 μm (single-side), thereby restricting improvements in energy density.

To overcome these limitations, dry electrode fabrication technologies that eliminate the use of liquid solvents have attracted considerable attention. In this process, active materials, conductive additives, and binders are dry-mixed and subsequently consolidated into freestanding electrode sheets through a calendaring process. Nevertheless, achieving a homogeneous distribution of conductive additives remains a significant challenge because conductive carbons with high specific surface areas and strong interparticle attractive forces tend to agglomerate during dry mixing. Although increasing the content or particle size of conductive additives has been explored to alleviate this issue, achieving uniform dispersion while maintaining low electrode resistance remains difficult. Therefore, the development of dry electrode manufacturing processes capable of improving conductive additive dispersion and suppressing electrode resistance is still required.

In this study, dry anodes for lithium iron phosphate (LFP) batteries were fabricated using artificial graphite and SiO_x as active materials, multi-walled carbon nanotubes (MWCNTs) and Super P as conductive additives, and polytetrafluoroethylene (PTFE) as a dry binder. The

compositions of the active materials, conductive additives, and binder were systematically varied, and the resulting electrodes were evaluated in terms of their physical and electrochemical properties.

The objective of this study is to establish the optimal composition and design criteria for high-performance dry anodes by investigating the effects of electrode composition on their physical and electrochemical characteristics. The findings are expected to improve conductive additive dispersion and reduce electrode resistance, while providing fundamental insights into the development of environmentally friendly, high-energy-density, and highly productive dry electrode manufacturing processes for next-generation LFP batteries.

Li-PAN/PAA Ionic-Conductive Binder Additive for Enhanced Cycling Stability and Rate Capability of Si/C Anodes and LFP Cathodes for LIB

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Silicon/carbon (Si/C) composite anodes offer high theoretical capacity but undergo severe volume expansion during lithiation, which disrupts conventional binder networks and accelerates capacity fading. Conventional binder systems such as PVDF or CMC/SBR provide either mechanical adhesion or elasticity, but lack sufficient ionic conductivity to sustain lithium-ion transport across the continuously fluctuating particle-binder interface. In this work, we present a Li-PAN/PAA ionic-conductive binder additive. Li-PAN provides polar nitrile groups and partially hydrolyzed ionic functionalities that facilitate Li^+ coordination and interfacial ion transport, while also contributing to the structural integrity of the electrode. Li-PAA, with its high density of lithium carboxylate ($-\text{COO}^-\text{Li}^+$) groups, forms strong interactions with silicon-based active materials, effectively accommodating repeated volume changes and maintaining electrode cohesion. Their complementary functions enable the Li-PAN/PAA composite binder to simultaneously improve mechanical stability and interfacial ion-transport kinetics. Coin half-cells fabricated with a CMC/SBR + Li-PAN/PAA hybrid binder applied to a Si/C anode delivered a discharge capacity of 514 mAh/g, an initial Coulombic efficiency (ICE) of 92%, and 92% capacity retention after 50 cycles at 1C, matching or exceeding the CMC/SBR-only baseline. Rate-capability testing further showed capacity retention of 95% at 5C and 85% at 7C, outperforming the CMC/SBR-only electrode and confirming the enhanced Li-ion transport enabled by the ionic-conductive network. We further extended the ionic-conductive binder concept to LFP cathodes using an SBR-free CMC/Li-PAN/PAA binder system, with evaluation focused on high-rate performance. Despite eliminating SBR from the formulation, the Li-PAN/PAA-modified LFP electrode retained 86% of its capacity at 5C and 75% at 7C, exceeding the rate capability of the conventional aqueous CMC/SBR binder. These results demonstrate that the Li-PAN/PAA binder platform improves cycling stability and rate capability across both anode and cathode materials, while also enabling a simplified, SBR-free binder formulation that maintains superior electrochemical performance, offering a practical route toward higher-performance next-generation lithium-ion batteries.

Structural Stabilization of Silicon Anodes Using Spray-Drying Si/C/CNT Composite

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Silicon (Si) has attracted considerable attention as a high-capacity anode material for next-generation lithium-ion batteries due to its high theoretical capacity. However, repeated volume changes during charge and discharge induce particle cracking and loss of electrical contact, while continuous formation of an unstable solid electrolyte interphase (SEI) leads to rapid capacity fading. In this study, Si/C/CNT composite anode materials were designed using a spray-drying process to improve the structural stability of silicon anodes. Fine Si particles were dispersed within a carbon matrix to accommodate the volume changes occurring during cycling, while carbon nanotubes (CNTs) were incorporated to form conductive networks within and between the composite particles. Secondary particles composed of Si, a carbon precursor, and CNTs were prepared by spray drying, and the effects of composition and synthesis conditions on particle morphology and microstructure were investigated. The relationship between the structural characteristics and electrochemical behavior of the prepared composites was analyzed to evaluate the effects of the Si/C/CNT composite structure on the structural stability and electrochemical performance of silicon anodes. This study demonstrates an effective approach to improving the stability of high-capacity silicon-based anode materials through the design of spray-dried Si/C/CNT composite structures.

Suppressing PTFE Reduction through Electron-Blocking Ion-Permeable Layer in Dry Anodes

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The transition toward solvent-free dry electrode manufacturing is a crucial step for reducing production costs and environmental impacts in next-generation lithium-ion batteries (LIBs). Polytetrafluoroethylene (PTFE) is widely recognized as an ideal polymeric binder for this process. Its unique fibrillation capability under mechanical shear stress enables the scalable fabrication of thick, high-area-loading electrodes without toxic solvents. However, deploying PTFE in graphite anodes is severely limited by its intrinsic electrochemical vulnerability. During initial lithiation, PTFE's low LUMO level makes it susceptible to severe reductive degradation specifically defluorination and carbonization. This irreversible breakdown depletes active lithium, forms highly insulating lithium fluoride (LiF), and destroys binder elasticity, ultimately causing drastically lower initial Coulombic efficiency (ICE) and rapid capacity fade over continuous cycling.

To overcome this critical barrier, we introduce a highly scalable, surface modification strategy: an electron-blocking ion-permeable layer (EBL). The optimized EBL has a thickness of approximately 8 nm and functions as a selective kinetic barrier. Its insulating nature cuts off electron tunneling pathways that trigger PTFE reduction, while its ether-rich molecular structure simultaneously facilitates rapid lithium-ion transport.

Comprehensive physical and electrochemical characterizations including XPS and Raman spectroscopy confirm the complete suppression of the typical PTFE reduction signature (0.6-0.7 V) and the robust preservation of the binder's structural integrity. When evaluated in practical NCM811||graphite full cells, the EBL-modified anode demonstrates a remarkable improvement in ICE, jumping from 76.75% to 83.19%, alongside an additional 16.50 mAh g⁻¹ of reversible capacity. Furthermore, the EBL-integrated cells exhibit superior long-term stability, maintaining 78.3% capacity retention after 500 cycles. Seamlessly integrating into commercial dry-mixing lines, this universal EBL design paves a highly practical pathway for stabilizing high-energy dry-processed anodes in advanced battery systems.

Development of a Metal-Oxide/Organic Hybrid Protective Layer for Dendrite-Free Lithium metal anode

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While lithium metal is considered an ideal anode material for next-generation, high-energy-density batteries, its commercialization is hindered by continuous dendrite growth and short cycle life. To overcome these challenges, this study developed a hybrid protective layer (HPL) composed of metal/metal oxide particles and organic materials to inhibit dendrite growth and facilitate lithium-ion transport. The protective layer utilizes a CCZ composite, incorporating multi-walled carbon nanotubes (MWCNTs) and copper oxide (CuO) for high electrical conductivity and mechanical strength, along with zinc oxide (ZnO) to enhance surface binding. This composite was blended with an organic polymer (PVDF-HFP) for flexibility and ionic conductivity, and coated onto the lithium metal. Successful synthesis was verified via EDS mapping and XRD analyses. Electrochemical evaluations demonstrated that the HPL-coated cell exhibited a resistance over 10 times lower than that of the bare cell in electrochemical impedance spectroscopy (EIS). Symmetric cell tests revealed a 1.3 times improvement in cycle lifespan and Coulombic efficiency. Furthermore, under a high C-rate condition of 5 mA cm⁻² and 1 mAh cm⁻², the HPL-coated cell recorded approximately 1.6 times lower resistance, demonstrating the exceptional stability and practical potential of this protective layer for high-performance lithium metal batteries.

Enhanced Electrochemical Performance of Over-Lithiated Layered Oxide Cathodes through Co Incorporation while Maintaining the Li-to-Transition-Metal Ratio

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Over-lithiated layered oxide (OLO) materials deliver higher specific capacities than conventional layered cathode materials through oxygen redox reactions occurring in the high-voltage region above 4.4 V during electrochemical cycling. In addition, their Mn-rich compositions offer advantages in terms of material cost, making OLOs promising cathode materials for next-generation lithium-ion batteries.

However, irreversible oxygen release and oxygen-vacancy formation during high-voltage cycling induce phase transitions from the layered structure to spinel- or rock-salt-like phases, resulting in severe voltage fading and capacity degradation. Furthermore, OLO materials inherently suffer from low electronic conductivity and sluggish interfacial reaction kinetics, which limit their rate capability and long-term cycling stability.

In this study, a small amount of Co was introduced during the calcination process while maintaining the Li-to-transition-metal ratio to improve the electrochemical performance of the OLO cathode material. The incorporation of Co can lower the energy barrier for oxygen extraction, thereby facilitating the electrochemical activation of the Li_2MnO_3 phase. In addition, Co introduction can improve the reaction kinetics and reduce the charge-transfer resistance.

Therefore, this study demonstrates that the simple introduction of a small amount of Co is an effective modification strategy for enhancing the electrochemical performance of OLO cathode materials.

Microstructural Regulation and Enhanced Lithium-Ion Transport in Dry-Processed Thick NCM Cathodes via BaTiO₃ Addition

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Dry-processed thick electrodes with high active-material contents and areal capacities have attracted considerable attention for the development of high-energy-density lithium-ion batteries. However, increasing the electrode thickness results in heterogeneous pore structures and extended lithium-ion transport pathways, leading to increased ionic resistance and degraded electrochemical performance, particularly at high charge–discharge rates. In this study, ferroelectric barium titanate (BaTiO₃, BTO) was introduced as an internal electrode additive to improve the microstructure and lithium-ion transport properties of dry-processed thick NCM cathodes. BTO was incorporated at concentrations ranging from 0 to 2.0 wt% into dry electrodes composed of NCM, Super P, and polytetrafluoroethylene, and the resulting structural and electrochemical properties were systematically investigated. Nano-computed tomography analysis revealed that the overall electrode porosity remained nearly constant at approximately 29%, whereas the addition of BTO reduced the tortuosity and improved the uniformity and connectivity of the pore and throat networks. In particular, the electrode containing 0.5 wt% BTO exhibited lower tortuosity and higher permeability than the BTO-free electrode, indicating the formation of effective lithium-ion transport pathways. Laser-induced breakdown spectroscopy further confirmed a more uniform lithium distribution through the electrode thickness after cycling. Among the investigated electrodes, the 0.5 wt% BTO-containing cathode exhibited the highest discharge capacity under high-rate conditions. This improvement is attributed to the regulation of the internal pore network and the facilitation of lithium-ion migration by local electric fields originating from the ferroelectric characteristics of BTO. In contrast, excessive BTO addition reduced the performance enhancement because of the increased amount of electrochemically inactive material and disruption of the electronic conduction pathways. These results demonstrate that the incorporation of an optimized amount of BTO can simultaneously improve the microstructure and ionic transport properties of dry-processed thick cathodes, providing an effective electrode design strategy for high-energy-density lithium-ion batteries.

Enhanced Rate Capability of LiFePO₄ Cathodes at Room and Low Temperatures through Ti Doping and Plasma Surface Modification

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Lithium iron phosphate (LiFePO₄, LFP) has a lower energy density than layered Ni-rich cathode materials such as NCM. Nevertheless, it has been widely adopted in electric vehicles owing to its excellent thermal stability, long cycle life, and structural robustness. With the increasing demand for energy storage systems operating under harsh environmental conditions, improving battery performance at low temperatures has become increasingly important. However, LFP intrinsically suffers from low electronic conductivity of approximately 10^{-9} S cm⁻¹ and sluggish Li-ion diffusivity of approximately 10^{-14} cm² s⁻¹ at room temperature. At low temperatures, reduced ionic mobility and increased internal resistance further aggravate these limitations, resulting in severe polarization and restricted high-power performance.

In this study, Ti doping combined with H₂/Ar plasma treatment was employed to improve the rate capability of LFP at both room and low temperatures. Ti⁴⁺ doping can modify the electronic structure and Li-ion transport characteristics of LFP, thereby improving its electrochemical reaction kinetics and high-rate performance at room temperature. In addition, H₂/Ar plasma treatment can selectively modify the particle surface. Highly reactive hydrogen species can partially remove or reduce oxidized Fe-containing surface species, while energetic Ar species can remove weakly bound surface contaminants and expose additional electrochemically active sites. These surface modifications are expected to reduce the activation barrier for interfacial charge transfer and suppress the substantial increase in polarization that occurs at low temperatures. Therefore, the combined application of Ti doping and H₂/Ar plasma treatment provides a complementary bulk- and surface-modification strategy for improving the rate performance of LFP over a wide operating-temperature range.

Enhanced Interfacial Adhesion and Thermal Stability of Aluminum Current Collectors for Dry-Processed NCM Cathodes via a P3HT-CNT-BaTiO₃ Primer Layer

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Dry-processed thick NCM cathodes have attracted considerable attention as a promising technology for high-energy-density lithium-ion batteries. However, the weak interfacial adhesion between the electrode and the current collector, resulting from the solvent-free process and low binder content, deteriorates electrochemical performance and thermal stability. In this study, a primer layer composed of poly(3-hexylthiophene) (P3HT), carbon nanotubes (CNTs), and barium titanate (BaTiO₃, BTO) was introduced onto an aluminum current collector to enhance interfacial stability. The effects of BTO content on the adhesion properties and electrochemical behavior were systematically investigated. The primer layer formed a uniform coating on the aluminum surface and increased the interfacial adhesion strength by more than five times compared with the bare aluminum current collector, as confirmed by SAICAS analysis. LIBS analysis revealed a more uniform lithium distribution after cycling, indicating improved ionic transport within the electrode. Furthermore, high-temperature electrochemical impedance spectroscopy demonstrated low interfacial resistance below 130 °C, whereas a sharp increase in resistance was observed at 140 °C owing to the suppression of the electronic conduction pathway. These results indicate that the P3HT-CNT-BTO primer layer effectively enhances interfacial adhesion while improving thermal safety by inhibiting electron transport at elevated temperatures. This work presents an effective interfacial engineering strategy for simultaneously improving the electrochemical performance and thermal stability of dry-processed thick electrodes for next-generation lithium-ion batteries.

Hollandite-type Vanadium Oxyhydroxide with High Energy Density as a Cathode Material for Lithium-Ion Batteries

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Herein, a nanosized hollandite-type VO_{1.75}(OH)_{0.5} is introduced as a high-energy cathode material for Li-ion batteries (LIBs). Rietveld refinement and bond-valence-energy landscape analysis based on X-ray diffraction reveal the Li⁺ diffusion pathways along the c-axis and possible atomic sizes of Li⁺ in the structure. VO_{1.75}(OH)_{0.5} exhibits a high discharge capacity of ≈ 300 mAh g⁻¹ (≈ 780 Wh kg⁻¹) at 0.05C, with an average operation voltage of 2.6 V (1.5–3.7 V vs. Li⁺/Li) based on reversible V⁴⁺/V³⁺ redox reaction. Even at 7C, it retains ≈ 195 mAh g⁻¹ (≈ 508 Wh kg⁻¹), $\approx 65\%$ of the capacity at 0.05C, with a capacity retention of $\approx 80\%$ over 200 cycles and small volume change (below $\sim 5.8\%$) upon discharge and charge. The lithium storage mechanism further reveals an outstanding high-power capability of ≈ 540 Wh kg⁻¹ (≈ 207 mA g⁻¹) with $\approx 80\%$ capacity retention over 300 cycles at 5C.

Keywords: Hollandite; Intercalation; Cathode material; Lithium ion Battery.

Understanding the Interfacial–Mechanical Degradation Feedback in High-Voltage Single-Crystal Mid-Ni Layered Oxide Cathodes

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Single-crystal mid-Ni layered oxides operated at high voltage represent a promising cathode platform, achieving energy densities comparable to those of Ni-rich cathodes while mitigating their intrinsic chemical and mechanical instabilities. Realizing their full potential, however, requires a mechanistic understanding of the multimodal degradation induced by high-voltage cycling. Here, we systematically investigate the stage-dependent degradation of single-crystal $\text{LiNi}_{0.6}\text{Co}_{0.1}\text{Mn}_{0.3}\text{O}_2$ (NCM613) cycled at 4.6 V through surface-to-bulk characterization spanning the initial, early, middle, and late stages of cycling. We show that cathode–electrolyte interphase (CEI) formation during the first cycle dominates early-stage capacity fade, whereas anisotropic strain and planar gliding remain largely reversible. With continued cycling, however, the CEI fractures under repeated volume changes, introducing non-uniform current distribution and additional mechanical stress that render planar gliding progressively irreversible. This establishes a coupled interfacial–mechanical degradation cycle: CEI fracture exposes fresh surfaces to electrolyte decomposition, while irreversible gliding generates persistent surface steps that sustain CEI regrowth. At the late stage, this coupling drives interfacial, crystallographic, and microstructural degradation to converge into a fully coupled state. These findings demonstrate that interfacial stabilization and mechanical tolerance must be addressed in concert—rather than independently—to unlock the full high-voltage potential of single-crystal mid-Ni cathodes.

Fluorinated Polymer Electrolytes with Enhanced Flame Retardancy and Wide-Temperature Electrochemical Performance

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The practical implementation of high-energy-density lithium-metal batteries requires not only high ionic conductivity and electrochemical stability but also the simultaneous mitigation of electrolyte flammability and low-temperature performance degradation. Conventional ether-based polymer electrolytes exhibit favorable lithium-ion transport properties; however, their high flammability and polymer-chain crystallization at low temperatures significantly reduce ionic conductivity and cell performance.

In this study, a flame-retardant polymer electrolyte based on a fluorinated polymer network was designed. The incorporation of fluorinated functional groups effectively suppressed combustion, and self-extinguishing time (SET) tests confirmed enhanced flame retardancy compared with the ether-based electrolyte. In addition, the disordered molecular arrangement inhibited polymer-chain packing and low-temperature crystallization, resulting in a high ionic conductivity of 1.73×10^{-4} S cm⁻¹ even at -40 °C.

The developed electrolyte exhibited stable oxidative behavior above 5 V in linear sweep voltammetry measurements. It also showed low polarization and stable voltage profiles during lithium plating/stripping tests, indicating that the formation of a stable electrode–electrolyte interface effectively suppressed nonuniform lithium deposition and interfacial side reactions.

Overall, the proposed fluorinated polymer electrolyte simultaneously overcomes the flammability and low-temperature crystallization limitations of conventional ether-based electrolytes, providing an effective design strategy for highly safe lithium-metal batteries capable of stable operation over a wide temperature range.

Synergistic Dual-Plasticizer Design for Balancing Ion Transport and Network Integrity in Gel Polymer Electrolytes

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Gel polymer electrolytes (GPEs) are promising for safer lithium metal batteries because they combine liquid-like ion transport with solid-like mechanical stability. However, conventional GPE design often focuses on modifying the crosslinker while overlooking the compatibility between the plasticizer and polymer network. Poor compatibility can result in incomplete crosslinking, structural heterogeneity, and unstable lithium-metal interfaces. Here, we introduce a dual-plasticizer strategy that regulates plasticizer–crosslinker affinity to balance ionic transport and network integrity. The electrolyte combines an ion-conducting ether plasticizer with an ester plasticizer that improves compatibility with an acrylate-based crosslinker. Hansen solubility parameter analysis and semiempirical calculations indicated stronger interactions between the acrylate-based crosslinker and the ester plasticizer than with the ether plasticizer. Consistent with these results, spectroscopic and X-ray scattering analyses confirmed more complete polymerization and the formation of a homogeneous three-dimensional network.

The resulting dual-plasticizer GPE retained favorable ionic conductivity while exhibiting improved tensile and compressive properties compared with its single-plasticizer counterpart. The homogeneous network promoted uniform lithium deposition and enhanced the cycling stability of both Li symmetric and LiFePO₄||Li cells. Stable operation at temperatures up to 120 °C and after sequential cutting and folding of pouch cells further demonstrated its wide-temperature operability and mechanical damage tolerance.

This work demonstrates that plasticizer–crosslinker affinity plays an important role in regulating network formation and electrochemical performance in GPEs. By connecting molecular compatibility with network uniformity, mechanical integrity, and interfacial stability, the proposed dual-plasticizer strategy provides a rational approach to developing safe and durable gel polymer electrolytes for lithium metal batteries.

Poster 26-022

Effect of Vacuum Sealing Conditions on Electrolyte Composition, Anode Interface, and Cycle Life in Lithium Metal Battery Pouch Cells

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Lithium metal batteries are widely regarded as promising candidates for high energy density beyond conventional lithium-ion systems, but their practical use remains limited by poor cycle life and non-uniform solid electrolyte interphase (SEI) formation. During pouch cell fabrication, the vacuum sealing step inevitably induces electrolyte splashing, which changes the residual electrolyte composition and directly affects cycle life. However, this phenomenon has not been studied in detail. In this study, we investigate the effects of vacuum sealing conditions on the residual electrolyte composition, anode interface, and cycle life of lithium metal pouch cells. First, we examine the trade-off in choosing the vacuum level, where a higher vacuum removes internal gas bubbles and suppresses bubble-induced non-uniform SEI growth but simultaneously intensifies electrolyte splashing and accelerates capacity decay. Second, we analyze the residual electrolyte recovered before and after sealing by NMR to reveal how the vacuum level and holding time govern the selective loss of solvent and salt and the resulting SEI composition. Lastly, we correlate these compositional changes with cycling behavior to identify an optimal sealing condition that balances bubble removal against electrolyte loss. Overall, we aim to show that control of vacuum sealing conditions is key to suppressing electrolyte splashing and improving the practical use of high-energy-density lithium metal pouch cells.

Enhancing the Electrochemical and thermal stability of Ultrahigh-Nickel $\text{LiNi}_{0.96}\text{Co}_{0.02}\text{Mn}_{0.02}\text{O}_2$ Cathodes via Mechano-fusion-Assisted Li-Zr-O surface modification

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Lithium-ion batteries (LIBs) are important energy-storage systems for electric vehicles and renewable energy applications because of their high energy density and long cycle life. In particular, ultra-high-Ni layered oxide cathodes ($\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$, $x \geq 0.9$) have attracted considerable attention due to their high discharge capacity ($>220 \text{ mAh g}^{-1}$). However, the high Ni content of these materials leads to severe interfacial instability, including surface reconstruction, oxygen release, lack of thermal stability, electrolyte decomposition, and the formation of inactive residual lithium compounds. These degradation phenomena accelerate impedance growth and capacity fading. Therefore, Surface coating has been widely employed to suppress direct contact between the cathode and electrolyte and to stabilize the highly reactive surface of ultra-high-Ni cathodes. Among various coating materials, Li-Zr-O-based surface layers have attracted considerable interest because they can provide chemical and structural protection without significantly hindering Li-ion transport. These surface layers can be formed through interfacial reactions between Zr-based coating materials and residual lithium compounds, such as LiOH and Li_2CO_3 , present on the surface of ultra-high-Ni cathodes. However, conventional wet-coating processes require large amounts of solvent, additional drying steps, and complicated process control, thereby increasing environmental burdens and the costs associated with solvent recovery and waste treatment. Moreover, wet-coating processes may suffer from particle agglomeration, uncontrolled precursor precipitation, and redistribution of coating species during solvent evaporation, potentially resulting in nonuniform surface coverage. In this study, $\text{LiNi}_{0.96}\text{Co}_{0.02}\text{Mn}_{0.02}\text{O}_2$ (NCM9622), an ultra-high-Ni layered oxide cathode containing 96 mol% Ni, was selected as a model material with particularly high surface reactivity and interfacial instability. A solvent-free mechano-fusion process was employed to uniformly distribute a Zr-based coating material over the cathode surface. Subsequent heat treatment promoted the reaction between the dry-coated Zr-based species and residual lithium compounds, resulting in the formation of a Li-Zr-O-based protective interphase. The effects of the dry-coating process and heat-treatment temperature on the surface chemistry, structural stability, and electrochemical performance of NCM9622 were systematically investigated.

An Al₂O₃/PSSLi-Coated Separator for Enhanced Li⁺ Transport and Long-Term Cycling Stability of Lithium Metal Batteries

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Polypropylene (PP) separators are widely used in lithium-metal batteries (LMBs) owing to their excellent mechanical properties. However, their poor electrolyte wettability and limited thermal stability hinder long-term electrochemical performance. In this study, a functional separator was developed by coating a bare PP separator with a thin (~3 μm) composite layer composed of Al₂O₃ nanoparticles and poly(lithium 4-styrenesulfonate) (PSSLi). Compared with the bare PP separator, the Al₂O₃/PSSLi separator exhibited significantly improved electrolyte wettability and thermal stability. Furthermore, the negatively charged sulfonate groups of PSSLi effectively suppressed anion transport, thereby promoting selective Li⁺ migration and increasing the lithium transference number from 0.49 to 0.55. The enhanced Li⁺ transport characteristics facilitated more homogeneous lithium-ion flux during cycling. Consequently, the capacity retention of the NCM811//Li cell employing the using Al₂O₃/PSSLi separator after 300 cycles at 1C was 74.7%, whereas that of the cell using the bare PP separator was 59.3%. The enhanced electrochemical performance is attributed to the synergistic effects of improved electrolyte affinity and selective Li⁺ transport induced by the Al₂O₃/PSSLi coating layer. These results demonstrate that the Al₂O₃/PSSLi separator provides an effective strategy for simultaneously improving the safety and long-term cycling stability of high-energy-density lithium-metal batteries.

Sublimation-Driven Pore Architecture in Dry-Processed Thick Cathodes for High-Rate, Long-Life Lithium-Ion Batteries

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Dry electrode processing offers a solvent-free route to high-area-capacity lithium-ion battery cathodes, yet the densely consolidated particle–binder network of dry thick electrodes impedes electrolyte penetration and through-thickness ionic transport, producing steep reaction gradients, severe polarization, and underutilization of the active material.

Herein, we demonstrate a transient pore-engineering strategy for dry-processed thick $\text{LiNi}_{0.9}\text{Co}_{0.05}\text{Mn}_{0.05}\text{O}_2$ cathodes using a residue-free sublimable additive that is completely removed by a mild thermal treatment, leaving distributed internal pores while preserving the fibrillated binder network and electrode integrity. At the optimal additive content, the electrode porosity increases from 24% to 39% and the electrolyte fully wets the electrode within 15 minutes, while symmetric-cell measurements reveal a $\sim 24\%$ reduction in ionic resistance. Depth-resolved laser-induced breakdown spectroscopy (LIBS) further visualizes markedly more uniform lithium extraction and insertion across the electrode thickness during 3C operation. Consequently, pore-engineered cathodes at $5\text{--}7\text{ mAh cm}^{-2}$ deliver higher capacities at high rates and sustain stable 1C cycling over 200 cycles, in contrast to the abrupt failure of dense counterparts near 180 cycles, whereas excessive additive contents degrade performance, defining a clear porosity optimization window between ionic transport and electronic/mechanical integrity.

These findings establish transient, sublimation-driven pore engineering as a practical material–process co-design route that reconciles electrode density with ionic transport in solvent-free, high-energy lithium-ion battery electrodes.

Keywords: #Lithium-ion batteries, #Dry process, #Thick electrode, #Pore structure engineering, # $\text{LiNi}_{0.9}\text{Co}_{0.05}\text{Mn}_{0.05}\text{O}_2$

Reactive Sulfur-Embedded Carbon Additives for Enhanced Cycling Stability and Thermal Safety of Oxygen-Releasing Layered Cathodes

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The safety of high-energy lithium-ion batteries is increasingly limited by the cathode itself: at high states of charge, Ni-rich and Li-rich layered oxides release lattice oxygen that oxidizes the electrolyte and triggers self-accelerating exothermic reactions culminating in thermal runaway. Conventional countermeasures, external thermal management and passive physical barriers, act only after considerable heat has already evolved, leaving the initiating chemistry unaddressed and motivating material-level strategies that intervene at the source.

Here, we introduce a sulfur-embedded porous carbon (S@C) as a multifunctional electrode additive that provides such an internally activated safeguard. The carbon framework immobilizes and homogeneously distributes active sulfur throughout the electrode while preserving electronic percolation. When incorporated into Li-rich layered oxide cathodes, the additive substantially improves cycling stability; for Ni-rich NCM cathodes, differential scanning calorimetry (DSC) of charged electrodes reveals mitigated exothermic heat flow, indicating enhanced intrinsic thermal stability. These improvements are consistent with the embedded sulfur serving as a reactive sink: near its low melting point, sulfur mobilizes within the electrode and combines with reactive oxygen species released from the delithiated lattice, interrupting oxygen-driven electrolyte decomposition at its onset.

By correlating electrochemical performance with thermal analysis, this work outlines a simple, scalable additive route to material-level safety and suggests reactive oxygen scavenging as a general design principle for inherently safer high-energy layered cathodes.

Keywords: #Lithium-ion batteries, #Thermal runaway, #Oxygen release, #Sulfur-carbon additive, #Layered oxide cathodes

Mitigating Mn Crossover and Interfacial Degradation in Sodium-Ion Batteries via a Multifunctional Ceramic-Coated Separator

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Sodium-ion batteries (SIBs) have emerged as a compelling alternative to lithium-ion systems for large-scale energy storage, primarily owing to the abundance and cost-effectiveness of sodium. However, the practical deployment of SIBs with transition-metal-based cathodes is frequently hindered by severe interfacial degradation, exacerbated by electrolyte decomposition and transition metal dissolution. Herein, we introduce a multifunctional ceramic-coated separator fabricated by applying polydopamine-functionalized ceramic nanoparticles onto a nonwoven polymer substrate. The functional ceramic coating layer significantly improves electrolyte wettability and Na⁺ transport, while effectively trapping dissolved Mn species to suppress their migration toward the hard-carbon anode. Therefore, the ceramic-coated separator mitigates Mn crossover and prevents anode degradation during cycling. The NaNi_{1/3}Fe_{1/3}Mn_{1/3}O₂||hard-carbon full cell with the ceramic-coated separator achieves 84.0% capacity retention after 100 cycles, compared with 54.1% for the cell with the bare separator. Post-cycling XPS analysis confirms substantial Mn accumulation on the ceramic-coated separator and reduced Mn deposition on the hard-carbon anode, supporting the suppression of Mn crossover. This study highlights the critical role of multifunctional separators in regulating Na⁺ transport and Mn migration, providing an effective strategy for improving the cycling stability of sodium-ion batteries.

Interfacial Chemo-Mechanical Degradation Behavior of Layered Oxide Cathodes in Sulfide- and Halide-Based All-Solid-State Batteries

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All-solid-state batteries, unlike conventional lithium-ion batteries based on liquid electrolytes, rely on solid–solid contacts between cathode active materials and solid electrolytes. Liquid electrolytes can readily infiltrate porous composite electrodes and maintain continuous ionic pathways even in the presence of local volume changes of the active material. In contrast, the solid–solid interfaces in all-solid-state lithium batteries are susceptible to electrochemically induced contact loss, local stress accumulation, interfacial side reactions, and the formation of resistive decomposition products.

Sulfide solid electrolytes are promising for all-solid-state batteries because of their high ionic conductivity and favorable mechanical deformability, which facilitate intimate contact with electrode materials. However, sulfide electrolytes possess a narrow electrochemical stability window and severe interfacial side reactions at high operating potentials. In comparison, halide

solid electrolytes have wide electrochemical stability window and provides high oxidative stability against high-voltage cathodes. Nevertheless, halide electrolytes exhibit lower ionic conductivity than sulfides and poor stability against lithium metal anodes.

In this study, $\text{Li}_6\text{PS}_5\text{Cl}$ (LPSCl), a representative sulfide solid electrolyte, and Li_3InCl_6 (LIC), a representative halide solid electrolyte, were individually applied to layered oxide cathode particles for dry surface modification. LPSCl and LIC were characterized in terms of their ionic conductivity, crystal structure, particle morphology, and mechanical properties. Sulfide based all-solid-state half cells were assembled to evaluate their charge–discharge performance, cycling stability, and the evolution of cathode–solid electrolyte interfacial resistance using electrochemical impedance spectroscopy.

Dual-Anion Locally Concentrated Ionic Liquid Electrolytes for Enhanced Ion Transport and Stable Interphase Formation in Thin Lithium Metal Batteries

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Ionic liquid electrolytes are attractive for safe lithium metal batteries because of their negligible volatility and high thermal and electrochemical stability, but their practical use is limited by high viscosity, low ionic conductivity, and poor interfacial wettability. Here, we report a dual-anion locally concentrated ionic liquid electrolyte (D-LCILE) containing FSI⁻ and TFSI⁻ anions with a fluorinated ether diluent. Classical molecular dynamics simulations using radial distribution functions and coordination-number analyses revealed that the dual-anion configuration reorganized the Li⁺ solvation environment and promoted aggregate-rich coordination. Ab initio molecular dynamics simulations further identified distinct anion dissociation behavior at the Li metal interface: FSI⁻ readily released fluorine species contributing to LiF formation, whereas TFSI⁻ exhibited higher structural stability. X-ray photoelectron spectroscopy, time-of-flight secondary ion mass spectrometry, and scanning electron microscopy confirmed the formation of a compact LiF-rich solid electrolyte interphase and dense, uniform lithium deposition. Electrochemical measurements showed improved separator wettability, an ionic conductivity of 1.053 mS cm⁻¹, a Li⁺ transference number of 0.879, and sufficient oxidative stability in linear sweep voltammetry. These bulk and interfacial advantages enabled stable Li plating/stripping and galvanostatic cycling of LiFePO₄ full cells using a 20 μm thin Li metal anode, achieving 99.93% capacity retention after 200 cycles at 1C. These results demonstrate that dual-anion solvation engineering can simultaneously enhance ion transport and interfacial stability in high-energy-density lithium metal batteries.

A Sustainable One-Pot Spray-Drying Strategy for Fabricating High-Capacity LiFePO₄ Thick Dry Electrode via Nanoparticle Reconstruction

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Lithium iron phosphate (LFP) is widely recognized as a highly safe and economical cathode material, essential for electric vehicles and large-scale energy storage. To satisfy the growing demand for higher energy densities, fabricating thick electrodes is critical. Yet, traditional slurry-cast manufacturing struggles to accommodate this. It relies on hazardous, costly NMP solvents requiring energy-intensive recovery systems. Furthermore, standard drying inevitably triggers binder migration, causing uneven internal structures that inflate costs and degrade the structural consistency, kinetics, and overall energy density.

To resolve these fundamental challenges, we introduce a sustainable, one-pot spray-drying hybrid method for producing high-performance LFP dry electrodes. The core innovation is a precisely regulated "Nanoparticle Reconstruction" mechanism. By utilizing pre-nanosized LFP—customized as precisely size-controlled powder clusters for optimal morphological manipulation—we substantially reduce solid-state lithium-ion diffusion distances while maximizing the active surface area.

The fabrication involves co-processing the LFP active material, fibrillizable PTFE binder, and SWCNTs as an advanced conductive scaffold within an entirely aqueous solution. During spray-drying, this dispersion is instantaneously atomized and dehydrated, yielding a composite precursor powder with a meticulously tailored nanoscale framework. The ultra-fast drying kinetics enable SWCNTs to construct a resilient 3D conductive web encapsulating the LFP particles, simultaneously ensuring an even, segregation-free distribution of PTFE.

This cutting-edge dry process eradicates the reliance on hazardous solvents and massive recovery infrastructures. Crucially, it completely suppresses binder migration, preserving exceptional mechanical durability, electrical connectivity, and structural stability even in ultra-thick electrode designs. Our research presents a highly scalable approach to boosting active material loading via refined conductive networks. This advanced dry-electrode methodology establishes a highly effective and eco-friendly framework for producing next-generation, high-energy-density LFP batteries

Electric-double-layer ordering couples dynamic anion replenishment with F-rich interphase formation in thin lithium metal batteries

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Thin lithium metal anodes are essential for realizing high-energy-density batteries, yet their practical implementation is hindered by rapid Li inventory loss caused by unstable and spatially heterogeneous interphase formation. Here, we demonstrate that a saturated lithium bis(fluorosulfonyl)imide (LiFSI)-coated separator functions as a localized salt reservoir that programs the electric double layer (EDL) at the Li metal interface. Operando Raman spectroscopy reveals that the salt-derived interfacial environment enriches anion-associated solvation structures, suppresses free-solvent accumulation, and dynamically replenishes FSI⁻ consumed during the initial stages of interphase formation. Polarization-dependent Raman measurements further indicate enhanced interfacial ion ordering, while potential-of-zero-charge analysis confirms substantial modification of interfacial charging behavior. Finite-element simulations, Li||Cu nucleation measurements, and impedance spectroscopy combined with distribution-of-relaxation-times analysis show that the ordered EDL homogenizes Li-ion flux, accelerates interfacial kinetics, and suppresses spatially nonuniform Li deposition. Molecular simulations and electronic-structure calculations indicate that AGG-rich Li⁺-FSI⁻ coordination favors anion-centered reduction. Consistently, ToF-SIMS depth profiling verifies the formation of a thin, uniform, F-rich solid-electrolyte interphase with reduced organic decomposition products. Consequently, LFP||Li full cells employing 20 μm thin-Li-metal anodes exhibit enhanced rate capability and stable cycling, retaining 91.9% of their initial capacity after 250 cycles at 0.5 C. These results establish localized-reservoir-driven EDL ordering as a molecular-level interfacial design principle for thin lithium metal batteries.

Nanochannel Orientation-Engineered Mesoporous Carbon Spheres for High-Power Potassium-Ion Hybrid Supercapacitors

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Abstract.

Facilitating access to internal active sites through mesopores is crucial for accelerating K^+ storage kinetics in carbon anodes and realizing high-power potassium-ion hybrid supercapacitors (PIHCs). In this study, mesoporous carbon spheres (MCS) with controlled nanochannel orientations were investigated to clarify the influence of internal pore accessibility on potassium-ion storage. Open-end MCS (*oe*-MCS) and closed-end MCS (*ce*-MCS) were prepared through a multiscale phase-separation strategy. *oe*-MCS and *ce*-MCS primarily differ in their internal mesopore accessibility, determined by the orientation of cylindrical nanochannels, allowing the effect of mesopore orientation on K^+ storage behavior to be systematically evaluated. Consequently, compared with *ce*-MCS, *oe*-MCS exhibited enhanced K^+ adsorption, a larger capacitive contribution, and improved specific capacity. A PIHC assembled using the *oe*-MCS anode achieved a maximum energy density of 103 Wh kg⁻¹ while maintaining a maximum power density of 12,300 W kg⁻¹. In addition, the device retained 86.1% of its capacity after 20,000 cycles at 2.0 A g⁻¹. These results demonstrate that increasing internal pore accessibility through nanochannel orientation enhances capacitive K^+ storage and ion-transport kinetics, providing an effective mesopore-design strategy for high-power PIHCs.

Sequential Dual-Interfacial Engineering of High-Nickel Cathodes with Li_3PO_4 and SWCNTs for high-performance lithium-ion batteries

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High-nickel layered oxide cathodes ($\text{Ni} \geq 96$) are promising materials for high-energy-density lithium-ion batteries owing to their high specific capacity. However, their practical application is limited by severe interfacial degradation, including electrolyte decomposition, transition-metal dissolution, surface reconstruction, and microcrack formation during repeated cycling. Inorganic surface coatings can suppress these parasitic reactions, but excessive surface coverage may hinder charge-transfer kinetics because of their limited electronic conductivity. Conductive carbon coatings improve electron transport but generally provide insufficient chemical protection against highly reactive high-nickel cathode surfaces.

Herein, we propose a sequential dual-interfacial engineering strategy for high-nickel cathodes by integrating Li_3PO_4 and single-walled carbon nanotubes (SWCNTs). A limited amount of Li_3PO_4 was intentionally introduced onto the cathode surface to provide chemical protection while avoiding excessive surface coverage. Subsequently, an SWCNT network was introduced across the cathode surface to establish interconnected electronic pathways and mechanically reinforce the interfacial architecture. This functionally differentiated architecture combines chemical stabilization by Li_3PO_4 with electronic and mechanical reinforcement by SWCNTs, thereby improving the electrochemical stability of the high-nickel cathode.

The dual-modified cathode exhibited enhanced electrochemical performance and cycling stability compared with the pristine cathode. These findings demonstrate that functionally differentiated interfacial engineering provides an effective strategy for stabilizing high-nickel cathodes while preserving their electrochemical performance.

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Sulfur-Assisted Pore Reconstruction in Dry-Processed Cathodes for Practical Lithium–Sulfur Batteries

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Abstract.

Solvent-free dry processing is attractive for fabricating high-loading sulfur cathodes, but mechanical densification can restrict electrolyte-accessible pores and cause non-uniform sulfur utilization. Here, we propose a sulfur-assisted pore-reconstruction strategy for dry-processed lithium–sulfur cathodes. Sulfur, Ketjenblack, and polytetrafluoroethylene were directly mixed and roll-pressed, allowing sulfur particles to temporarily occupy internal domains within the consolidated electrode. Subsequent thermal treatment redistributed sulfur into the conductive carbon framework and generated additional pore space at the original sulfur-particle positions. In contrast to conventional electrodes prepared from preformed sulfur–carbon composites, the resulting electrode retained improved electrolyte-accessible pathways after densification. This reconstructed pore architecture promoted electrolyte infiltration and more uniform sulfur conversion under high-loading and lean-electrolyte conditions. These results demonstrate that active-material redistribution can be used to engineer transport pathways in dense dry electrodes without additional sacrificial templates or solvent-based processing.

Aspect-Ratio-Driven CNT Networks for Freestanding High-Energy Lithium–Sulfur Cathodes

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Abstract.

Freestanding sulfur cathodes can reduce inactive electrode components and improve the practical energy density of lithium–sulfur batteries. However, the structural requirements that allow carbon nanotube (CNT) networks to maintain mechanical integrity without binders or current collectors remain unclear. In this study, we investigated the influence of CNT geometry on the formation of self-supporting CNT–sulfur electrodes. Coarse-grained molecular dynamics simulations showed that long, thin CNTs form interconnected entanglement junctions that distribute mechanical stress while preserving porous transport pathways. In contrast, CNTs with lower aspect ratios preferentially formed localized bundles and mechanically fragile structures. A transition to stable freestanding networks was observed at a CNT aspect ratio of approximately 400. Guided by this geometric criterion, binder-free sulfur cathodes containing up to 90 wt% sulfur were fabricated using commercially available CNTs. The optimized electrode delivered 1,579.8 mAh g⁻¹ at 0.1 C and 578.9 mAh g⁻¹ at 5 C. Furthermore, a 1 Ah-class pouch cell operated under lean-electrolyte conditions achieved a gravimetric energy density of 430 Wh kg⁻¹. These results demonstrate that CNT aspect ratio provides a practical structural guideline for designing lightweight, mechanically robust, and high-energy sulfur cathodes.

Dry-Processed Double-layer Silicon/Graphite Anodes with Optimized Silicon-rich Layer Architecture for Enhanced Lithium-ion Transport

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Silicon (Si)-graphite composite anodes have attracted significant attention as promising next-generation anode materials for improving the energy density of lithium-ion batteries due to their high theoretical capacity. However, silicon undergoes a large volume expansion of over 300% during charge/discharge processes, resulting in electrode cracking, loss of electrical contact, and repeated formation of unstable solid electrolyte interphase (SEI) layers, which are major causes of rapid capacity fading and poor cycling stability. Therefore, electrode structural design strategies that can effectively mitigate the mechanical degradation of silicon while maintaining high-rate performance are highly required.

Dry electrode processing enables the fabrication of high-loading electrodes without the use of solvents. In particular, the fibrillation of PTFE binders forms a three-dimensional fibrous network that enhances the mechanical binding between active materials and effectively suppresses the repetitive volume changes of silicon. In addition, unlike wet electrode processes, dry processing minimizes binder migration caused by solvent evaporation and drying, allowing independent control of layer composition and binder distribution. This structural flexibility enables the design of various multilayer electrode architectures while maintaining uniform electrode structures even under high-loading conditions.

In this study, a double-layer silicon-graphite composite anode consisting of a silicon-containing layer and a graphite layer was fabricated by utilizing the structural design flexibility of dry electrode processing. The position of the silicon-containing layer was controlled toward either the separator-facing side or the current collector-facing side to investigate the effects of layer arrangement on lithium-ion reaction behavior. Furthermore, the electrochemical characteristics and reaction mechanisms of the layered electrodes were evaluated by comparison with a single-layer electrode containing the same silicon content.

Rate capability evaluation demonstrated that the position of the silicon-containing layer influenced the high-rate performance at 5C, which was associated with changes in lithium-ion accessibility and charge-transfer pathways within the electrode. To understand the origin of these electrochemical differences, EIS analysis was performed to investigate interfacial resistance and charge-transfer kinetics, and dQ/dV analysis was conducted to analyze the effects of the layered architecture on lithium insertion/deinsertion behavior and polarization characteristics.

Sn/Sb Co-doped $\text{Li}_6\text{PS}_5\text{Cl}$ Solid Electrolytes: Composition Design Using Bayesian Optimization

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$\text{Li}_6\text{PS}_5\text{Cl}$ (LPSCl) has attracted considerable attention as a sulfide solid electrolyte for all-solid-state batteries because of its high room-temperature ionic conductivity and favorable mechanical properties. Various P-site doping strategies have been investigated to further improve the performance of LPSCl, and both Sn and Sb have been reported to enhance Li ion transport and structural stability. However, rational optimization of multicomponent doping compositions remains challenging because complex interactions among dopants give rise to intricate composition–property relationships.

In this study, a Sn/Sb co-doping strategy was combined with Bayesian optimization to identify the optimal dopant composition for LPSCl. Sn/Sb co-doped LPSCl was synthesized from SnS_2 and Sb_2S_3 precursors through high-energy dry ball milling followed by heat treatment. The composition–property relationship was analyzed based on iterative synthesis and ionic conductivity measurements, enabling identification of the optimal composition region.

The optimization identified the composition region exhibiting the highest ionic conductivity and clarified the influence of Sn/Sb composition on ionic transport behavior. The optimized composition will be further validated through structural characterization, air-stability assessment, and electrochemical performance evaluation, while the underlying enhancement mechanism will be investigated using advanced structural analyses and computational approaches. This work presents a composition–design strategy that integrates Sn/Sb co-doping with Bayesian optimization and provides a foundation for efficient composition optimization of multicomponent sulfide solid electrolytes.

Simultaneous Control of Phase Purity and Grain-Boundary Chemistry in Mixed-Halide Argyrodite Solid Electrolytes

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Abstract.

Sulfide-based argyrodite solid electrolytes are promising for all-solid-state batteries because of their high Li⁺ ionic conductivity and good mechanical deformability. Mixed Cl/Br compositions can enhance bulk ionic conductivity by increasing anion disorder and improving Li⁺ migration pathways. However, overall Li⁺ transport also depends on grain boundaries, where residual phases and local halide composition can strongly affect ion conduction. In this study, we hypothesized that precursor particle size controls phase formation and halide distribution during solution-based synthesis. Fine Li₂S, LiCl, and LiBr precursors promoted argyrodite formation, reduced unreacted LiCl/LiBr phases, and enhanced Cl and Br diffusion toward particle surfaces. This produced a highly pure argyrodite phase with grain boundaries having a higher Cl/Br content than the bulk and faster Li⁺ transport. As a result, the electrolyte achieved an ionic conductivity of approximately 9 mS cm⁻¹ and improved all-solid-state battery performance. These results show that precursor size control can regulate both phase formation and grain-boundary chemistry in mixed-halide argyrodites.

Development of Cathode Materials for Sodium-Ion Batteries Using Multi-Electron Redox Reactions of Iron Cations

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Lithium-ion batteries (LIBs), currently in widespread use, rely heavily on rare metals, raising supply chain concerns as demand continues to grow. Therefore, to achieve a sustainable society, research on sodium-ion batteries (SIBs) utilizing earth-abundant sodium as a charge carrier is rapidly advancing. Sodium iron oxides are particularly promising cathode materials because iron is a low-cost, resource-abundant redox-active element. However, because sodium has a higher atomic weight than lithium, achieving a competitive energy density remains a critical challenge.

To address this issue, cathode materials capable of undergoing multi-electron redox reactions—which allow for greater electron transfer per unit mass—have attracted significant attention. Although iron can adopt a wide range of oxidation states up to Fe^{6+} states above Fe^{4+} are typically unstable "unusually high valence states," making the reversible utilization of multi-electron reactions inherently difficult. To overcome this, combining iron cationic redox with oxygen anion redox to achieve high capacity has been explored; however, challenges such as voltage hysteresis and structural instability due to oxygen gas evolution remain unresolved.

Therefore, to realize a stable, high-energy-density cathode, this study focuses on multi-electron reactions driven by iron cationic redox rather than oxygen anionic redox. Unusually high valence states such as Fe^{4+} to Fe^{6+} can be stabilized via strong orbital hybridization with oxygen.¹⁾ While conventional research has predominantly focused on octahedrally coordinated compounds, this study focuses on tetrahedral materials, in which Fe–O orbital hybridization is even stronger. Stable phases of sodium-rich iron oxides, including Na_3FeO_4 , $\text{Na}_3\text{Fe}_2\text{O}_7$, and Na_3FeO_3 , feature structures composed of isolated tetrahedra, corner-sharing dimers, and corner-sharing 1D chains, respectively. In this presentation, we evaluate these three sodium-rich iron oxides as SIB cathodes and discuss the feasibility and reaction mechanisms of reversible multi-electron redox at their tetrahedral iron sites.

Improved Cathode Utilization of Nanostructured VO₂(B) for Magnesium Rechargeable Batteries

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Magnesium rechargeable batteries (MRBs) are promising post-lithium-ion batteries because of their high volumetric capacity, resource abundance, and safety. However, the strong electrostatic interaction between Mg²⁺ and the anion framework leads to sluggish Mg²⁺ diffusion, resulting in poor cathode utilization at room temperature. Brookite-type vanadium oxide VO₂(B), which possesses a two-dimensional tunnel structure for Mg²⁺ diffusion, is a promising cathode material for MRBs^{[1][2]}. In this study, VO₂(B) nanoparticles were prepared by a solvothermal method to shorten the Mg²⁺ diffusion path and improve cathode utilization.

X-ray diffraction confirmed the formation of the VO₂(B) phase for both hydrothermal and solvothermal synthesis, while TEM observations revealed a reduction in particle size for the solvothermal sample. Electrochemical measurements showed that the nanoparticle sample delivered a higher discharge capacity than the hydrothermal sample. SEM/EDS analyses before and after cycling further revealed that a larger fraction of the discharge capacity originated from reversible Mg insertion/extraction, demonstrating improved utilization of the active material.

To clarify the structural reaction mechanism, XRD and V K-edge XAFS measurements were performed at different charge and discharge states. The diffraction peaks and absorption edge shifted reversibly during Mg insertion and extraction, and the same reversible behavior was maintained in the second cycle, indicating highly reversible structural and electronic changes. In addition, only a small change in the unit-cell volume was observed during cycling despite the high reversible capacity, suggesting excellent structural stability. These results demonstrate that nanosized VO₂(B) is a highly promising cathode material for MRBs, combining high cathode utilization, reversible Mg insertion/extraction, and minimal volume change.

Reference

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Multilayer Stacking for High-Loading Performance in Dry-Processed Si/Graphite Anodes

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Abstract.

High-loading Si/graphite anodes are promising for increasing the energy density of lithium-ion batteries, but thick dry-processed electrodes often suffer from nonuniform conductive networks and limited active-material utilization under high-rate conditions.

In this study, we present a simple multilayer stacking strategy to improve the structural and electrochemical properties of dry-processed thick Si/graphite anodes. CNT and Super P were used as representative fibrous and particulate conductive additives, respectively, to investigate the effect of conductive-additive distribution within the electrode architecture.

The multilayer electrodes exhibited denser structures than single-layer electrodes, while ML-C/C maintained a tortuosity comparable to that of SL-C despite its lower porosity. This result indicates that porosity or ionic tortuosity alone is insufficient to account for the electrochemical behavior of thick dry electrodes. Instead, ML-C/C showed the lowest volume resistivity and impedance among the representative electrodes, leading to improved rate capability. At 0.5C, ML-C/C delivered a discharge capacity of 238.92 mAh g⁻¹, whereas ML-C/S and SL-C delivered 205.74 and 64.51 mAh g⁻¹, respectively. Depth-resolved LIBS analysis further revealed a reduced Li signal gradient across the ML-C/C electrode thickness after lithiation, suggesting more consistent lithiation across the electrode depth.

Furthermore, the ML-C/C anode showed stable cycling behavior in NCM811||Si/graphite full cells and was further demonstrated in ultra-high-loading coin-type cells and 3042-format pouch-type cells. Overall, multilayer stacking provides a practical route to improving high-loading dry-processed Si/graphite anodes, addressing conductive-network formation, lithiation consistency, and cell-level applicability simultaneously.

Improved Electrochemical Performance of Ni-rich NCM Cathodes by Phosphorus-Based Coating

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The escalating demand for electric vehicles and large-scale energy storage systems has accelerated the development of high energy density and long cycle life lithium-ion batteries. Among various cathode materials, $\text{LiNi}_x\text{Co}_y\text{Mn}_z\text{O}_2$ (NCM) cathodes have attracted considerable attention due to their high specific capacity, excellent energy density, and reduced cobalt content. In particular, Ni-rich NCM cathodes are recognized as promising candidates for next-generation power sources.

However, Ni-rich NCM cathodes still suffer from several critical issues during prolonged charge/discharge cycling, including surface degradation, transition metal dissolution, intergranular microcracks, and structural instability. Furthermore, undesirable side reactions between the cathode surface and electrolyte accelerate capacity fading and deteriorate long-term cycling stability. These issues significantly limit the practical application of high-energy-density lithium-ion batteries. To overcome these challenges, various strategies such as surface coating, elemental doping, concentration-gradient structures, and electrolyte optimization have been extensively investigated.

Among these approaches, phosphorus-based surface modification has attracted considerable attention because phosphorus-containing species can form stable surface layers and suppress undesirable interfacial reactions with the electrolyte. Therefore, in this study, a phosphorus-based coating layer was introduced to enhance the interfacial stability and electrochemical performance of Ni-rich cathodes. As a result, although the coated cathode exhibited a slight decrease in initial discharge capacity, remarkable improvements in cycling efficiency, capacity retention, and long-term electrochemical stability were achieved. These findings demonstrate that P-based surface coating is an effective strategy for improving the durability of next-generation Ni-rich cathodes.

Enhancing Through-Thickness Ion Transport by Incorporating Soluble Li Salt into Dry-Processed Thick Cathodes

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Abstract.

dry-processed thick high-Ni NCM cathodes are promising for achieving high-energy-density lithium-ion batteries, but their performance is often constrained by ionic transport limitations and poor electrolyte infiltration. These bottlenecks lead to non-uniform reactions, increased polarization, and limited high-rate capability, emphasizing the need for scalable microstructure-engineering strategies to enhance electrolyte accessibility and ionic transport across thick cathodes.

Here, we introduce a soluble-additive incorporation strategy for dry-processed thick cathodes. Soluble additives are embedded in the electrode through a simple powder mixing process and subsequently released from the electrode matrix during electrolyte wetting. Their release generates additional electrolyte accessible pore area and ion transport pathways within the thick electrode.

This approach modifies the internal electrode structure without requiring substantial changes to the conventional dry-electrode fabrication process. Compared with the bare cathode, the additive containing cathodes exhibited improved high-rate capability and cycling performance, indicating that additive release alleviates ionic transport limitations in thick cathodes.

Structural and electrochemical analyses further revealed improved electrolyte infiltration, reduced ionic resistance, and more homogeneous electrochemical reactions across the electrode thickness. These results demonstrate that incorporating soluble additives into dry-processed thick cathodes provide a practical and scalable approach to improving through-thickness ion transport and electrochemical performance.

Impact of Process-Induced Iron Contamination on the Long-Term Cycling Degradation of Ah-Level NCM811/Graphite Pouch Full Cells

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Lithium-ion batteries are manufactured through sequential mixing, coating, calendaring, slitting, and notching steps, during which metallic particles can be introduced from the wear of stainless steel processing machinery. Iron is among the metallic species encountered in this way, and its behavior inside the cell is governed by the potential window it experiences. Metallic Fe is thermodynamically unstable at the operating potential of a charged layered oxide cathode and therefore undergoes oxidative dissolution. The resulting Fe ions migrate through the electrolyte and across the separator, and are reduced back to the metallic state at the low potential of the graphite anode. Such deposits act as sites for continued electrolyte reduction and solid electrolyte interphase reformation, which consumes cyclable lithium. Their localized growth can further establish a conductive path that manifests as a micro short circuit, accompanied by elevated self-discharge and reduced coulombic efficiency. These consequences are magnified in the Ni-rich $\text{LiNi}_{0.8}\text{Co}_{0.1}\text{Mn}_{0.1}\text{O}_2$ and graphite configuration, the most widely adopted system for high-energy-density lithium-ion batteries, whose narrow surface stability margin and strong sensitivity to lithium inventory loss leave little tolerance for any additional parasitic pathway. How the level of iron contamination translates quantitatively into full-cell performance loss, however, has not been resolved.

In this study, 1 Ah-class full cells were fabricated with Fe metal powder deliberately introduced into the NCM811 cathode slurry at 0, 1, 10, and 50 ppm, and accelerated degradation testing was conducted at 45 °C. After 900 cycles, the capacity retention decreased monotonically with increasing Fe content, from 63.9% for the pristine cell to 60.2, 58.4, and 57.4% for the 1, 10, and 50 ppm cells, respectively. The incremental loss was largest upon the initial introduction of Fe and progressively diminished at higher contamination levels, indicating that the degradation does not scale in direct proportion to the total Fe inventory. To identify the origin of this behavior, post-mortem SEM and EDS analyses were performed on the harvested electrodes to resolve the spatial distribution of Fe-related species and to correlate it with interfacial degradation of the

cathode and anode. Thermal stability analysis together with overcharge, forced discharge, and external short circuit tests was then carried out to assess whether the accumulated degradation alters the failure tolerance of the cell. The electrochemical, morphological, and safety results are discussed together to clarify how the Fe contamination level governs both the cycle life and the failure response of Ni-rich full cells.

SnS₂ Anchored on CNT Frameworks for Enhanced Polysulfide Adsorption and Redox Catalysis in Lithium–Sulfur Batteries

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Lithium–sulfur (Li–S) batteries have attracted considerable attention as next-generation energy storage systems owing to sulfur’s high theoretical capacity and natural abundance. However, their practical application remains hindered by the dissolution and migration of lithium polysulfides (LiPSs), which induce the shuttle effect, as well as by the sluggish kinetics of the multistep conversion reactions among sulfur species.

In this study, a catalytic SnS₂–CNT composite framework was designed in which nanoparticulate SnS₂ acts as an active catalytic host rather than merely serving as a conductive additive. The SnS₂/CNT composite was synthesized through a room-temperature stirring process, followed by mechanical milling and mild thermal annealing to establish a mechanically robust and electrically interconnected conductive network.

The polar surface of SnS₂ strongly adsorbs LiPSs through Lewis acid–base interactions, thereby effectively suppressing polysulfide diffusion and migration. In addition, SnS₂ provides electrocatalytically active sites that promote the reversible conversion of sulfur species and accelerate redox kinetics during charge and discharge.

The nanoscale morphology of SnS₂ facilitates intimate interfacial contact with the CNT network while exposing abundant active sites for LiPS adsorption and catalytic conversion. Consequently, the SnS₂–CNT hybrid framework enhances the reversibility of sulfur redox reactions and improves the overall electrochemical performance.

Furthermore, key synthesis parameters, including reaction time, precursor ratio, and the SnS₂-to-carbon ratio, were found to significantly influence particle morphology, dispersion, and catalytic activity. Ongoing studies are focused on heteroatom doping and CNT surface modification to further strengthen interfacial interactions and improve long-term cycling stability.

Optimization of a Transfer-Printed Al₂O₃-Rich Quasi-Solid-State Polymer Electrolyte for Lithium-Ion Batteries

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Abstract.

Quasi-solid-state polymer electrolytes have attracted considerable attention for semi-solid-state batteries by immobilizing liquid electrolytes within a crosslinked polymer matrix, thereby reducing electrolyte leakage and thermal risks. However, their relatively low mechanical strength and interfacial side reactions arising from residual unreacted monomers may compromise long-term battery stability and safety.

Herein, we report a transfer-printing strategy for the fabrication of an ultrathin Al₂O₃-rich composite electrolyte layer on graphite anodes. The polydopamine coating on the Al₂O₃ surface effectively improves the dispersibility of the Al₂O₃ particles. Through an optimized transfer-printing process, a thin and uniform composite film with an average thickness of 13 μ m was successfully fabricated on graphite anode. The transferred layer was subsequently infiltrated with a precursor solution containing a crosslinker and a liquid electrolyte, followed by in situ thermal crosslinking to form an Al₂O₃-rich quasi-solid polymer electrolyte.

The optimized transfer-printed electrolyte layer provided intimate interfacial contact with the graphite anode and reduced the electrode – electrolyte interfacial resistance. In addition, the incorporation of polydopamine-coated Al₂O₃ enhanced the thermal stability of the composite electrolyte. These results demonstrate that transfer printing is an effective processing strategy for integrating ceramic-rich quasi-solid-state polymer electrolytes with graphite anodes and may contribute to the development of safer, high-energy-density lithium-ion batteries.

Bi-Layer Solid Electrolyte Design for All-Solid-State Li-Metal Batteries

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Sulfide-based solid electrolytes (SSEs) are highly promising for all-solid-state Li metal batteries (ASSLBs), in that they allow Li metal anodes to be used without a flammable liquid electrolyte, offering a substantial gain in energy density. In such cells, however, a single electrolyte layer must simultaneously withstand the oxidizing cathode and the strongly reducing Li metal. Argyrodite $\text{Li}_6\text{PS}_5\text{Cl}$ (LPSCl), the most widely adopted SSE, is thermodynamically unstable against Li and forms a resistive interphase at the anode that raises the internal resistance of the cell. Decoupling these two opposing requirements into separate electrolyte layers is therefore a rational design strategy for ASSLBs.

In this study, we report a bi-layer SSE in which a dual halide doped $\text{Li}_7\text{P}_2\text{S}_8\text{I}_{0.5}\text{Br}_{0.5}$ (LPSIBr) layer, expected to be more compatible with Li metal¹, is placed adjacent to the anode while LPSCl is retained on the cathode side. NCM622|LPSCl|LPSIBr|Li and NCM622|LPSCl|Li cells were compared at an identical total SSE loading. Both delivered comparable capacities from 0.1 to 1.0 C, whereas after 100 cycles at 0.3 C the bi-layer cell retained approximately 95.8% of its capacity against approximately 87.6% for the single-layer cell. The Coulombic efficiency remained near 99.9% for both, indicating that the faster capacity fade of the single-layer cell reflects a growing internal resistance rather than the loss of cyclable Li. As the two cells differ only in the electrolyte adjacent to Li, this resistance is anticipated to occur at the anode-electrolyte interface. Guided by this, the growth of each resistance contribution is identified by time-resolved impedance analysis. This work shows that anode-side electrolyte selection, rather than modification of a single electrolyte, is an effective route to suppressing interfacial resistance growth in ASSLBs.

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Effect of Reduction Degree on the Electrochemical Performance of Magnesiothermally Reduced SiOx Anodes for Lithium-Ion batteries

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Silicon-based anode materials have been extensively investigated owing to their high theoretical capacities. However, the practical application of elemental Si remains limited by severe volume expansion during repeated lithiation and delithiation, leading to particle pulverization, loss of electrical contact, and rapid capacity fading. Silicon oxide (SiOx) has therefore attracted considerable attention owing to its high theoretical capacity of approximately 2500 mAh/g and better cycling stability than Si. The improved stability of SiOx is generally attributed to the formation of electrochemically inactive lithium-containing phases during the initial lithiation process, which act as a buffering matrix and alleviate structural degradation. Nevertheless, the electrochemical behavior of SiOx strongly depends on its oxygen content and reduction degree, making precise control of the reduction state essential for balancing specific capacity and cycling stability.

In this study, SiOx particles were synthesized by magnesiothermic reduction (MRR) of sol-gel-derived SiO₂ nanoparticles. The reduction temperature was systematically varied to control the oxygen content and crystallinity of SiOx. Structural and chemical analyses revealed that increasing the reduction temperature promoted the formation of crystalline Si while decreasing the oxygen content. Electrochemical evaluation demonstrated that increasing the reduction temperature promoted the formation of crystalline Si and reduced the oxygen content, resulting in higher discharge capacities but progressively poorer cycling stability owing to increased volume expansion, whereas cycling stability gradually deteriorated owing to larger volume changes during repeated lithiation and delithiation. The SiOx reduced at 800 °C exhibited the highest specific capacity of 1769.9 mAh/g. This study establishes reduction-degree control as a key design parameter for balancing capacity and cycling stability in SiOx anodes.

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Direct Growth of CNT Networks on ZnO Anodes via Rotary Chemical Vapor Deposition for Enhanced Lithium Storage

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The growing demand for high-energy-density lithium-ion batteries has stimulated extensive research into alternative anode materials beyond conventional graphite. Among the various candidates, zinc oxide (ZnO) has attracted considerable attention owing to its high theoretical capacity of 987 mAh/g, which is approximately 2.6 times higher than that of graphite.

However, the conversion and alloying reactions of ZnO are accompanied by substantial volume changes during repeated lithiation and delithiation, resulting in structural degradation and poor cycling stability. In addition, its intrinsically low electrical conductivity limits charge transport and practical electrochemical performance. Although carbon nanotubes (CNTs) have been incorporated to address these limitations, conventional physical mixing often leads to CNT agglomeration, non-uniform distribution, and insufficient interfacial contact.

In this study, CNT/ZnO composite powders were synthesized by directly growing CNT on ZnO particles using a rotary chemical vapor deposition (RCVD) process. Continuous reactor rotation enabled uniform CNT growth over the ZnO surface, forming an interconnected conductive network with enhanced interfacial contact. The synthesis conditions were systematically optimized, and the electrochemical performance of the composites was evaluated as lithium-ion battery anodes. The directly grown CNT network significantly improved electronic conductivity and structural integrity, leading to enhanced lithium-storage performance compared with pristine ZnO and physically mixed counterparts. This work demonstrates that direct CNT growth via RCVD is an effective strategy for constructing robust conductive networks in ZnO anodes and improving their electrochemical performance.

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Controlling Lithium Stripping Homogeneity via Depth-Of-Discharge Modulation during Formation

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Abstract.

Lithium metal batteries (LMBs) are a leading candidate for next-generation high-energy storage, yet their practical operation under low N/P conditions remains limited by the poor reversibility of the lithium metal anode during repeated plating and stripping. In discharge-initiated LMBs, where the cell is first discharged (stripped) before plating, interfacial non-uniformity generated during this initial stripping step can affect the subsequent plating morphology and cycle life. However, how the formation conditions—in particular the depth-of-discharge (DOD) - influence this initial interfacial behavior remains to be clarified, and a rational formation protocol for regulating it has yet to be established.

In this presentation, we introduce a DOD-controlled formation protocol for discharge-initiated LMBs that suppresses initial surface roughening and enhances long-term cycling performance. Combining morphological (Optical Microscopy, FIB-SEM) and electrochemical (overpotential, EIS) analyses with full-cell testing, we examine how the DOD during formation affects the uniformity of the stripping reaction and the resulting surface evolution, and how these in turn influence the subsequent plating and cycling behavior.

This early-stage surface control improves subsequent plating uniformity and stabilizes long-term cycling. Notably, these benefits are achieved by tuning the DOD during formation, without any structural or interfacial modification, providing a practical design guideline for discharge-initiated LMBs.

A Gel-Rich Hybrid Sulfide Solid Electrolyte for Stable Low-Pressure and Low-Temperature All-Solid-State Batteries

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Abstract.

All-solid-state batteries (ASSBs) have emerged as promising next-generation energy storage systems owing to their high energy density and enhanced safety. Among various solid electrolytes, sulfide solid electrolytes have attracted considerable attention because of their high ionic conductivity. However, conventional sulfide electrolyte membranes consisting of inorganic particles and polymer binders form rigid electrode–electrolyte interfaces, resulting in poor interfacial contact and high interfacial resistance, particularly under low-pressure and low-temperature operating conditions. Herein, we propose a hybrid sulfide solid electrolyte membrane by incorporating an ionic liquid-based gel polymer into a $\text{Li}_6\text{PS}_5\text{Cl}$ (LPSCI) framework to construct a compliant and ion-conductive interfacial architecture. During membrane fabrication, a gel-rich interfacial layer is spontaneously formed at the electrode–electrolyte interface, improving interfacial contact and accommodating local deformation. This unique structure facilitates homogeneous Li^+ transport, suppresses interfacial polarization, and reduces interfacial impedance during cycling. Consequently, the hybrid membrane delivers stable electrochemical performance under low stack pressure and low-temperature conditions, where conventional sulfide electrolyte membranes typically suffer from severe interfacial degradation. These findings demonstrate that gel-rich interfacial engineering is an effective strategy for overcoming the intrinsic interfacial limitations of sulfide solid electrolytes, providing a practical design principle for high-performance all-solid-state batteries.

Roll to Roll Processable, UV Curable Composite Polymer Electrolytes for Room Temperature

Solid State Batteries

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Solid state batteries are promising alternatives to conventional liquid electrolyte systems. In this study, we developed a UV curable composite polymer electrolyte based on polybutadiene urethane acrylate (PBDUA), propylene carbonate (PC), lithium bis(fluorosulfonyl)imide (LiFSI), and $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO). Rapid UV curing enabled flexible electrolyte membranes to form within seconds and provided a route for continuous roll to roll processing.

Before large area fabrication, the PBDUA to LiFSI mass ratio was varied from 10:1 to 10:9 to optimize the composition. All compositions exhibited room temperature ionic conductivities on the order of $10^{-4} \text{ S cm}^{-1}$, with the 10:3 composition showing the highest value. However, higher salt concentrations provided improved oxidation and reduction stability and more stable lithium plating and stripping behavior. FTIR analysis showed that increasing LiFSI content decreased the contributions from free PC and uncoordinated PBDUA carbonyl groups, while increasing those from Li^+ coordinated carbonyl groups. Changes in the FSI^- related band also indicated a salt rich coordination environment. Considering conductivity, electrochemical stability, and lithium metal compatibility, the 10:9 composition was selected for scale up.

LLZO was incorporated to improve selective lithium ion transport, increasing the lithium ion transference number from 0.345 to 0.795. The electrolyte was then coated onto a thin PVdF support and cured by UV irradiation. The support suppressed pore formation caused by curing shrinkage and enabled a mechanically stable large area membrane. The resulting

membrane exhibited an ionic conductivity of $1.87 \times 10^{-4} \text{ S cm}^{-1}$ at room temperature and an electrochemical stability window of approximately 1.5 to 4.1 V. Lithium symmetric cells showed stable cycling for more than 300 cycles at 0.1 mA cm^{-2} , while LFP half cells maintained stable performance for more than 100 cycles at 0.5 C. These results demonstrate a scalable strategy for room temperature solid state lithium batteries.

Mechanical Stress Regulation of Initial Lithium Stripping to Suppress Pitting in Discharge-Initiated Lithium Metal Batteries

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For practical applications, operating at a low negative-to-positive (N/P) ratio is essential to maximize energy density. However, lithium metal batteries (LMBs) are exceptionally sensitive to such low N/P conditions. Under these regimes, interfacial non-uniformity during lithium stripping severely accelerates cell degradation, with surface destruction being particularly severe in discharge-initiated cells. While external pressure is known to enhance long-term cycle performance, its specific role in mitigating initial pitting remains poorly understood. In this study, we investigate the impact of mechanical stress on initial lithium stripping to establish an optimal pressure for suppressing surface roughness. Using confocal laser scanning microscopy (CLSM), we quantified 3D morphological changes—including pit distribution and area fraction—under varying applied pressures. Furthermore, focused ion beam (FIB) cross-sectional analysis was employed to track how these pressure-controlled initial surfaces influence subsequent lithium plating. Our results demonstrate that optimal external stress disperses localized stripping, reduces pit dimensions, and mitigates overall surface roughening. This physical regulation stabilizes overpotential behavior and significantly improves active lithium retention. Ultimately, optimizing applied pressure prevents the morphological collapse of the lithium surface, providing critical mechanical guidelines for practical LMB design.

Fabrication of High-Areal-Capacity Thick LiFePO_4 Electrodes by Stacking Free-Standing Electrode Sheets

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Growing demand for longer driving ranges in electric vehicles has driven the development of advanced electrode design strategies for high-energy-density lithium-ion batteries. Thick electrodes are particularly promising because they enable high active-material mass loading while reducing the relative fraction of inactive components, such as current collectors and separators.

However, conventional slurry-based processing of thick electrodes often causes conductive additives and binders to migrate toward the electrode surface during drying. The resulting compositional inhomogeneity can lead to crack formation, poor adhesion, and reduced mechanical stability. In addition, poly(vinylidene fluoride) generally requires N-methyl-2-pyrrolidone as a processing solvent, raising environmental and manufacturing concerns. Solvent-free dry-electrode processing has therefore been investigated as an alternative; however, homogeneous component dispersion, coating uniformity, and crack suppression remain challenging.

In this study, we propose a fabrication strategy for thick lithium iron phosphate (LFP) electrodes based on free-standing electrode sheets while retaining compatibility with conventional slurry-based electrode processing. LFP thick electrodes with areal capacities ranging from 4 to 11 mAh cm^{-2} were successfully fabricated. The proposed electrode architecture alleviated compositional migration and crack formation associated with increased electrode thickness while enabling high areal capacity. Electrochemical performance was evaluated using galvanostatic charge–discharge cycling, rate-capability testing, and electrochemical impedance spectroscopy. An LFP|Li half-cell employing an electrode with an areal capacity of 7.5 mAh cm^{-2} exhibited a capacity retention of 98% after 30 cycles at a current density of 0.38 mA cm^{-2} . Furthermore, variations in electrode porosity with electrode composition were analyzed to elucidate the effects of conductive-additive and binder contents on the electrode structure.

These results demonstrate a practical electrode fabrication strategy for achieving high mass loading and high areal capacity, offering a promising approach toward high-energy-density lithium-ion batteries.

Discontinuous Carbon Nanofiber Bundle–Fluoropolymer Composite Interlayer for Regulating Lithium Deposition toward Anode-Free Lithium Metal Batteries

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Anode-free lithium metal batteries offer high theoretical energy density by eliminating the need for a preloaded Li metal anode. However, because their operation relies on a limited Li inventory supplied exclusively by the cathode, nonuniform Li deposition and the continuous formation of electrochemically inactive Li lead to rapid capacity decay. Here, we introduce a composite interlayer composed of electrospun three-dimensional carbon nanofiber (CNF) bundles and fluorinated polymers, including poly(vinylidene fluoride) (PVDF) and poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP). The combined effects of low-temperature carbonization and the discontinuous CNF bundle architecture reduce the effective electronic conductivity of the interlayer, thereby regulating local electron transport and promoting bottom-up Li deposition from the Cu/interlayer interface. In addition, the quasi-network structure formed by the intertwined CNF bundles and polymer binder mechanically suppresses deposition-induced volume expansion and facilitates dense Li deposition. The incorporated β -phase-rich fluoropolymer components endow the interlayer with lithiophilicity and promote the formation of an F-rich solid electrolyte interphase, thereby stabilizing the electrode/electrolyte interface. Morphological analysis confirmed that the composite interlayer retained its network structure without discernible collapse after prolonged cycling, and the observed Li morphology suggested that the interlayer regulated Li deposition. Compared with bare Cu, interlayer-coated Cu exhibited a 53.9% lower initial Li nucleation overpotential. In half-cell measurements, bare Cu exhibited rapid degradation after 80 cycles, whereas interlayer-coated Cu maintained stable Li plating and stripping for more than 200 cycles, with average Coulombic efficiencies of 93.5% and 98%, respectively. Moreover, symmetric cells incorporating the composite interlayer maintained stable cycling for 800 h, approximately twice the lifetime of the control cells. These results demonstrate that the combined regulation of electron transport, Li deposition behavior, and interfacial chemistry enabled by the composite interlayer effectively mitigates irreversible Li loss, providing an interfacial design strategy relevant to the development of anode-free lithium metal batteries.

Direct Growth of CNT Networks on LiFePO_4 Cathodes for High-Performance Lithium-Ion Batteries

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Lithium iron phosphate (LiFePO_4 , LFP) has attracted considerable attention as a promising cathode material for lithium-ion batteries owing to its low cost, excellent thermal stability, long cycle life. Nevertheless, its practical application, particularly under high-rate and low-temperature operating conditions, remains limited by intrinsically low electronic conductivity and kinetically limited lithium-ion diffusion within the olivine lattice. Conventional strategies for overcoming these limitations generally involve carbon coating, conductive additive incorporation, particle-size reduction, or hybridization with carbon nanotubes (CNTs). However, the mixing of LFP particles and conductive additives often results in nonuniform conductive networks, localized agglomeration, and insufficient interfacial contact, particularly in solvent-free dry electrode fabrication.

In this study, CNTs were directly grown on the surface of LFP particles using a rotary chemical vapor deposition process to construct a homogeneous and interconnected conductive network. The resulting LFP and CNT composites were applied to dry-processed cathodes without using N-methyl-2-pyrrolidone-based organic solvents. Directly grown CNTs on LFP's surface effectively connected adjacent LFP particles and provided intimate interfacial contact with the active material, thereby facilitating electron transport and improving the utilization of LFP. The optimized cathode delivered specific discharge capacities of 166.3 mAh/g at 0.1 C and 134 mAh/g at 2 C. These results demonstrate that direct CNT growth on LFP particles is an effective approach for improving conductive-network uniformity and electrochemical performance, while also providing a promising strategy for environmentally sustainable and cost-effective dry electrode manufacturing.

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Engineering precursor intermediates as coating and doping platforms

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Surface coating and doping have become key strategies for stabilizing high-energy-density layered oxide cathodes. To date, these modifications have primarily targeted two material states: hydroxide precursors and lithiated cathode materials, leaving oxide intermediate states unexplored as an additional processing window for precise cathode interfacial engineering.

Here, we introduce a precursor intermediate as a new platform for coating and doping. We demonstrate that the hydroxide/oxide phase fraction and porosity of the precursor intermediate can be precisely tuned through controlled heat treatment. These intermediate characteristics govern the activation energy of the subsequent lithiation reaction, enabling controlled lithiation of oxide intermediates. As a proof of concept, Zr was introduced through the precursor intermediate, demonstrating sequential coating, surface doping, and interfacial phase engineering within a single processing window. Cross-sectional STEM–EDS/EELS analyses together with XPS depth profiling reveal the spatial distribution and chemical state of Zr, demonstrating that the precursor-intermediate strategy provides two new opportunities for cathode interfacial engineering through the independent control of interfacial chemistry and internal accessibility.

Granule-Assisted Transport Engineering of High-Loading Graphite Anodes for Solvent-Free Lithium-Ion Batteries

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Dry processing offers a promising route toward high-energy lithium-ion batteries by eliminating solvent-removal steps and enabling thick-electrode fabrication. However, graphite anodes remain difficult to implement through conventional dry-processing approaches because binder distribution, particle alignment, and ionic transport become increasingly critical at high areal loading. Here, we demonstrate a granule-assisted electrode design in which graphite, carbon black, and a conventional CMC/SBR binder system are integrated into composite secondary particles through spray drying. Rapid droplet contraction reorganizes platelet-shaped graphite into three-dimensional granules with multidirectional particle orientation and increased edge-plane exposure. Following solvent-free thermal roll pressing, the resulting electrodes exhibit a nearly uniform porosity profile across the electrode thickness and markedly reduced binder segregation compared with slurry-cast graphite electrodes.

These structural features promote more homogeneous lithiation and alleviate transport polarization as electrode loading increases. At an areal capacity of 6.9 mAh cm^{-2} , the granule-based electrode preserves distinct graphite staging behavior and exhibits improved Li-ion transport characteristics relative to its slurry-processed counterpart. The transport advantage is further retained in practical full cells employing dry-processed NCM811 cathodes, where the granule-based anode delivers improved high-rate capability and 81.8% capacity retention after 200 cycles at 1C, compared with 71.5% for the conventional graphite electrode. These results highlight particle-scale architectural control as an effective strategy for translating conventional graphite chemistry into high-loading, solvent-free electrode manufacturing.

Engineering hydrolytic stability and reversible O3/P3/OP2 transitions in O3-type sodium-ion battery cathodes through Cu²⁺ doping

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O3-type layered oxides have emerged as promising cathode materials for sodium-ion batteries (SIBs) owing to their competitive energy density and structural robustness. However, their inherent sensitivity to moisture remains a critical obstacle for practical application. In this study, we introduced Cu²⁺ doping into NaNi_{0.3}Fe_{0.2}Mn_{0.5}O₂ (NFM325) to synthesize NaNi_{0.3}Fe_{0.2}Cu_{0.1}Mn_{0.4}O₂ (NFCM3214). Comprehensive analyses (XRD, FTIR, TGA, XPS, SEM, and TEM) demonstrate that Cu incorporation prevents bulk degradation and suppresses surface reactions such as Na⁺/H⁺ exchange and the formation of NaOH and Mn hydroxides upon water exposure. These local structural modifications enhance both bulk and surface stability against moisture. Electrochemical evaluations confirm that NFCM3214 retains 89% of its initial capacity after direct water contact, with significantly improved Na⁺ diffusivity, rate capability (7.2 → 88.8 mAh g⁻¹ at 700 mA g⁻¹), and cycling retention (71.4% → 84.5% after 100 cycles). Mechanistic insights further reveal that Cu²⁺ doping mitigates Jahn–Teller distortion, promotes the formation of an OP2 intermediate phase during deep desodiation, and facilitates reversible OP2–O/P transitions during cycling. Finally, full-cell tests using an N-doped MoS₂ anode demonstrate high capacity and stable cycling performance, underscoring the practical potential of Cu doped O3-type layered oxides for advanced sodium-ion batteries.

Resolving the High-Voltage Trilemma in Solid-State Batteries via All-Component Crosslinked Polymer Electrolytes

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Driven by the escalating energy density requirements of electric vehicles and wearable electronics, traditional liquid lithium-ion batteries are rapidly approaching their physicochemical limits. Their widespread application is fundamentally hindered by uncontrolled interfacial side reactions, dendritic lithium propagation, and irreversible active material loss, which collectively compromise safety and cycling stability. While solid polymer electrolytes (SPEs) present a promising paradigm shift toward intrinsically safer solid-state lithium metal batteries (SSLMBs), mitigating the inherent trade-off between room-temperature ionic conductivity and high-voltage oxidative stability remains a formidable challenge.

Herein, we report a highly integrated, diazide-based all-component crosslinked SPE synthesized via a rapid UV photocuring process. During photolysis, highly reactive nitrene intermediates facilitate non-selective C–H insertion, covalently locking fluorinated elastomers (PVDF-HFP), ionic liquids (EMIM-TFSI), plasticizers (succinonitrile), and lithium salts (LiTFSI) into a singular, robust polymer network. We designed custom fluorinated crosslinkers—ethane-1,2-diylbis(4-azido-2,3,5,6 tetrafluorobenzoate) (2Bx) and its ethylene oxide-extended counterpart (2Bx-4EO): whose strong electron-withdrawing effects substantially depress the highest occupied molecular orbital (HOMO), thereby intrinsically bolstering anodic stability. Furthermore, systematically modulating the crosslinker chain length enables the precise architectural tuning of continuous lithium-ion transport channels. The incorporation of a trace fluoroethylene carbonate (FEC) additive further passivates the lithium metal anode, suppressing deleterious parasitic reactions. Crucially, the liquid precursor demonstrates excellent compatibility with scalable manufacturing via direct blade-coating onto electrodes. Ultimately, this comprehensive design strategy successfully resolves the trilemma of high-voltage stability, mechanical flexibility, and interfacial compatibility, paving a viable route for next generation high-energy SSLMBs.

Key Words: Solid polymer electrolyte (SPE); Solid-state lithium metal batteries (SSLMB); UV photocuring; Diazide crosslinker; High-voltage stability; Nitrene intermediate