Cobalt-Free Cathodes for Next Generation Lithium-Ion Batteries

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NEXCERIS where energy meets environment

Project ID: bat417

OVERVIEW

Timeline

- Project Start Date: Jan. 2019
- Project End Date: Mar. 2022
- Percent Complete: ~ 66 %

Barrier and Technical Targets

- Cycle Life: 1000 cycles C/3 deep discharge with < 20 % energy fade
- Cost: < $100/kWh

Budget

- Total Project Funding: $3.08 M
  - DOE share: $2.46 M
  - Cost share: $620 k
- Funding for FY2020: $1.1 M
- Funding for FY2021: $1.0 M

Partners

- Ohio State University: Dr. Jung-Hyun Kim
  Battery testing
  Cell chemistry development
- Navitas Systems Dr. James Dong
  Large-scale electrode fabrication
  2-Ahr battery manufacture and testing

RELEVANCE

Impact

- The increased demand for EVs is driving demand for battery materials.
- Renewed interest in reduced/cobalt free Li-ion battery cathode formulations
  - Opportunity to reestablish U.S. dominance in batteries

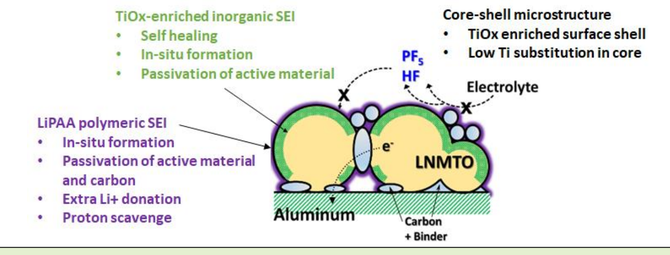
Objective

- Develop high performance and cobalt-free Li-ion battery based on high voltage lithium manganese nickel titanium oxide (LNMTO) cathode and complementary cell chemistry (electrolyte/cathode formulation)
- Identify low-cost, scalable process for producing cathode powder.
- Identify strategy for Nexceris to grow domestic manufacturing and create jobs to support new clean energy and e-mobility opportunities

APPROACH

Develop cobalt-free cell based on high-voltage $LiNi_{0.5}Mn_{1.2}Ti_{0.3}O_4$ (LNMTO) cathode

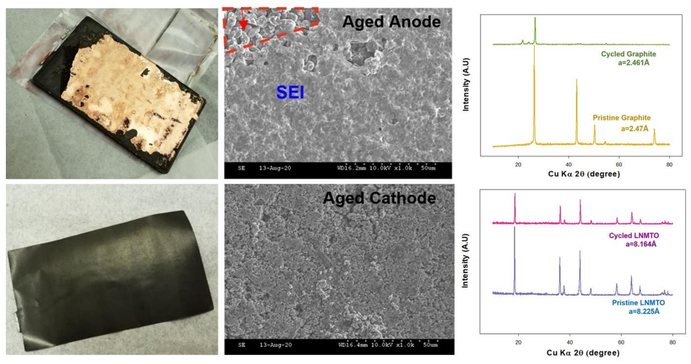
- Improve cycle and calendar life by forming a solid-electrolyte interface that effectively passivates the cathode surface
- Create novel LNMTO core/shell microstructures where Ti is preferentially located at surface and partner with optimized binder/electrolyte chemistries



ACCOMPLISHMENT: PPC Post-mortem analysis

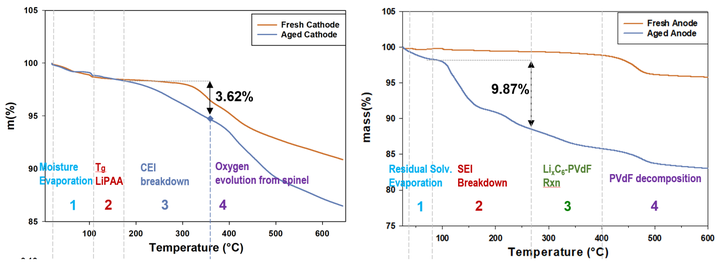
Completed comprehensive post-mortem analysis of tested 2-Ah PPC cells

- No electrolyte found during cell teardown – severe electrolyte decomposition
- Graphite anode delaminated from Cu current-collector, adhered separator
- Transition metal dissolution from cathode – migration to anode



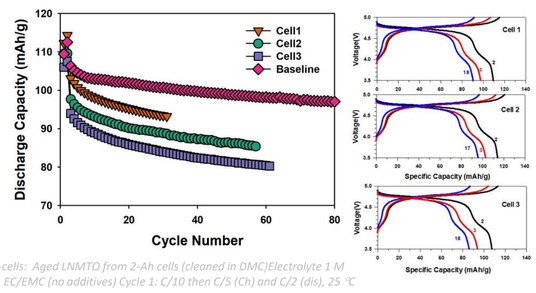
TGA analysis completed to quantify the CEI and SEI layers

- Confirmed SEM – heavily deposited SEI layer ~ 10 wt.% of aged anode



Aged 2-Ah cells recovered and prepared as coin half-cells

- Aged cathodes deliver full charging/discharging capacity – same as pristine
- Inferior capacity retentions due to large cell impedances from CEI



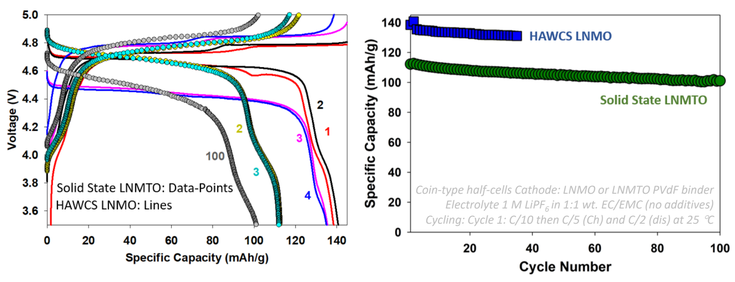
Coin-type half-cells: Aged LNMTO from 2-Ah cells (cleaned in DMC)Electrolyte 1 M $LiPF_6$ in 1:1 wt. EC/EMC (no additives) Cycle 1: C/10 then C/5 (Ch) and C/2 (dis), 25 ℃

ACCOMPLISHMENT: *HAWCS* Powder Synthesis

Developed alternative low-cost powder synthesis approach to scale production

- Two-step Hybrid Alternative Wet-Chemical Synthesis (*HAWCS*) process
- Enables excellent compositional and particle morphology control
- Scalable, modified precipitation process without strict process controls

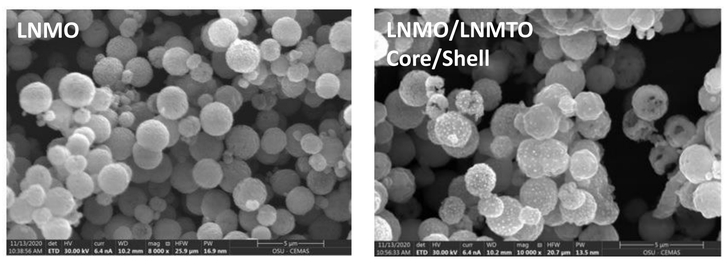
Enhanced cell performance demonstrated compared to Y1 solid-state powder



In parallel core/shell hierarchical powder microstructure concept advanced

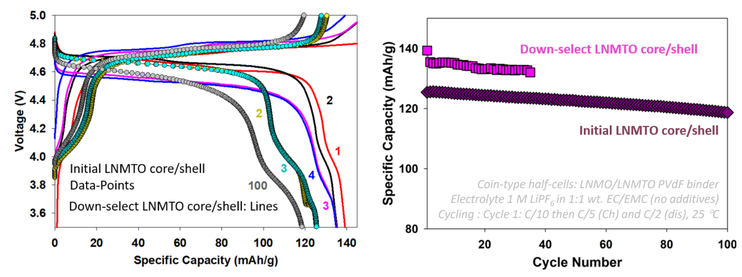
- Identified most appropriate deposition approach and shell thickness
- Integrated shell process with HAWCS core-powder production process

High purity LNMO/LNMTO spherical powder successfully produced



Optimization of the core/shell process

- Significant improvement in initial capacity –retain core LNMO performance

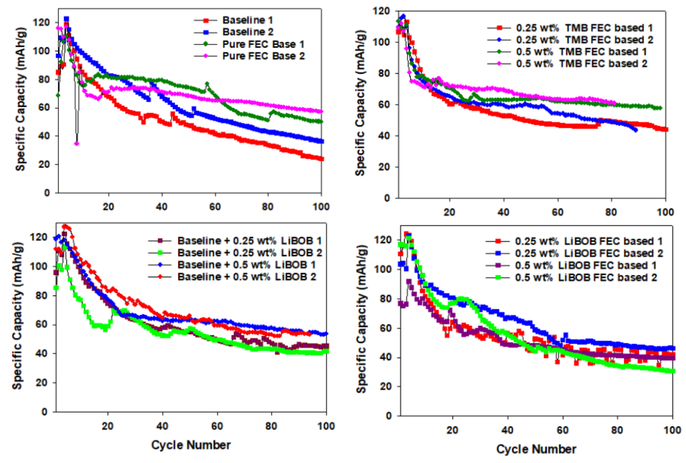


ACCOMPLISHMENT: SLP/2-Ah Cell Development

The team has focused more on single-layer pouch (SLP) and additional 2-Ah cell testing to better understand cell-chemistry interactions

Electrolyte study completed at SLP scale to try and identify a complementary electrolyte for the LNMO/LNMTO core-shell cathode

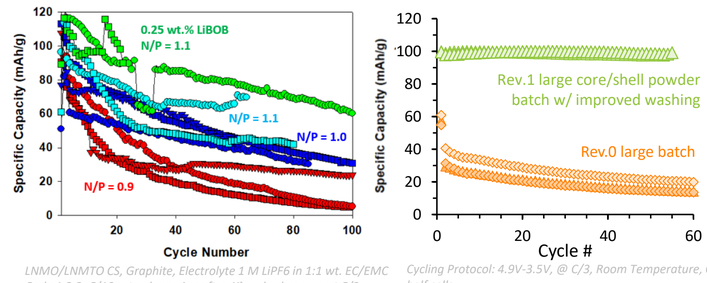
- Working to identify opportunities to collaborate with other teams, and leverage their expertise to help deconvolute cathode/electrolyte



LNMO/LNMTO CS, LiPAA binder, Graphite, Electrolyte Baseline 1 M LiPF6 in 1:1 wt. EC/EMC, Pure FEC based: 1 M LiPF6 in 1:4 FEC/EMC Cycle 1,2,3: C/10 rate, degassing after 1st cycle charge, rest C/3 (cells cycled at C/5-Ch and C/2-Dis for first 10 cycles

Identifying optimal n/p ratio and cathode formulation

Large *HAWCS* powder batches created new issues that had to be addressed through process improvements to lithiation and washing processes



LNMO/LNMTO CS, Graphite, Electrolyte 1 M LiPF6 in 1:1 wt. EC/EMC Cycle 1,2,3: C/10 rate, degassing after 1st cycle charge, rest C/3

Cycling Protocol: 4.9V-3.5V, @ C/3, Room Temperature, Coin half cells

Cell chemistry, formation cycle and scaled-up LNMO/LNMTO core-shell powder down-selected and 2-Ah cells fabricated and tested

- Initial capacity improved versus PPCs
- Cycle life did not demonstrate improvement – learnings from the test incorporated into future work (i.e., need to increase powder particle-size)

MILESTONE REVIEW

Milestone	Metric	Status
Post-Mortem analysis completed on PPCs **(M14)**	Characterization of PPCs post completed and strategies to address top degradation mechanisms defined	**Completed** ○ Post-mortem characterization of PPCs and SLP completed, and used to inform development work
Core-shell LNMTO cathode powder down-Select **(M18)**	Promising core-shell LNMTO powders down-selected that improve cell performance compared to the homogeneous LNMTO powder	**Completed** ○ New (HAWCS) process refined and down-selected ○ Core-shell approach integrated with HAWCS process ○ Best-in-class half-cell performance demonstrated
Cell chemistry Down Select **(M21)**	Promising cell chemistries down-selected based on SLP cell testing	**Completed** ○ Cell-chemistry finalized ○ Formation cycle condition down-selected
Core-shell based LNMTO cathodes demonstration **(M24)**	Intermediate 2 Ah cells with down-select core-shell LNMTO cathode and cell chemistry	**Completed** ○ 2-Ah cells and tested ○ Initial capacity better than Year 1 ○ Disappointing cycle-life, plan identified to address

REMAINING CHALLENGES AND BARRIERS

This last year has been difficult. There are several remaining challenges with the proposed future work is focused on addressing.

End-User OEM Interest

- Nexceris has talked with auto-OEMs and stakeholders throughout the value-chain, including cathode powder manufacturers. This customer discovery has helped inform Nexceris strategy and where it can provide most-value.

Cathode Powder scale-up

- Realizing challenges with co-precipitation, pivoted and developed HAWCS process. Again, there was difficulty in scaling batch size for 2-Ah cells. Need to focus on larger-batch sizes and address delta in performance.

Identification of complementary cell chemistry (electrolyte)

- A literature review and electrolyte study has been completed but electrolyte degradation is still an issue and barrier to improving cell performance.

RESPONSES TO REVIEWERS' COMMENTS

We appreciate the reviewer feedback from our 2020 AMR presentation. Main concerns which we have worked to address include:

1. *"initial results show that LNMTO cathode capacity is decreased while the cyclability is not improved to meet the project targets.", "performance of LNMO control material is very poor"*

Initial performance has been improved in FY21, but cycle life is primary concern to achieve project targets.

2. *"scale-up that does not show the same cycling performance"*

This has continued to be a challenge – we are focusing on larger batch sizes

PROPOSED FUTURE WORK

Future work is focused on addressing the challenges identified in FY21 and meeting the project targets.

- Strategy changed to complete additional 2-Ah cell iterations
- Deconvolute powder scale-up/chemistry issues.

Future Work	Description
Post-mortem analysis on FY21 2-Ah cells Ensure address primary degradation mechanism	○ Mitigation strategies for gas-generation ○ Help identify better electrolyte formulation
Tune LNMO powder particle size Increase particle size for high V system	○ Process control to target 20 µm particle size ○ Re-target shell coating process
Powder batch size optimization Equivalent performance with scale-up	○ Segmentation of powder process
Electrolyte/Cathode Formulation Optimization Identify complementary cell chemistry	○ Both internal/external work to identify appropriate cell chemistry to pair with cathode

COLLABORATIONS/COORDINATION OTHER INSTITUTIONS

Nexceris is fortunate to have excellent project partners that have supported cell chemistry development, testing and large cell (2-Ah) manufacture and testing.

Nexceris has developed a very productive relationship with ANL and looks forward to building more relationships and collaborations in the future.

Project Team Member	Relationship
The Ohio State University	○ Coin-cell and SLP cell screening of cell chemistries ○ Cell chemistry (additives/binder) development ○ Analytical characterization of cathode materials and electrodes
Navitas Systems	○ Electrode scale-up, formation-cycling optimization ○ Large format 2-Ahr battery fabrication and testing

SUMMARY

Accomplishments

- Identified and implemented new synthesis process for producing cathode powders that provides greater microstructural control and is easier to scale than precipitation process.
- Incorporated coating process to produce LNMO/LNMTO core/shell powder
- Completed manufacturing and testing of intermediate 2-Ah cells – informs strategy to improve performance (esp. cycle life).
- Detailed post-mortem characterization of PPCs completed.

Impact towards VTO Objectives

- HAWCS provides a scalable and flexible synthesis process to support custom development – provides Nexceris with a unique synthesis capability.
- Quality of the LNMO powder has been improved – need to identify and incorporate complementary electrolyte chemistry to achieve cycle life.

This poster does not contain any proprietary, confidential, or otherwise restricted information. Any proposed future work is subject to change based on funding levels