High-Performance Electrolyte for Lithium-Nickel-Manganese Oxide (LNMO)/Lithium-Titanate (LTO) Batteries

Project ID: bat 441 • 2021 AMR Review

Principal Investigator/Presenter: Dr. Jennifer Hoffmann for Gotion, Inc.

Timeline

- **Start of Project:** March 20, 2019
- **End of Project:** March 30, 2022*
  *As of March 2021, when approved project extension was received
- **Status:** ~ 60% Completed

Budget

- **Total Project Funding:** $2.63 Million
  - **DOE:** $1.27M
  - **Industry:** $1.36M

Partners

- **University of Rhode Island** (URI): Analytical Support and Additive synthesis

Overview

Month/Year	Description of Milestone or Go/No Go Decision	Status
Oct. 2020	Of developed additives, determine which should be scaled up for large scale testing	Completed
Jan. 2021	Submit for no cost extension/project revision due to impact of COVID-19 and other delays	Completed
Sept. 2021 (Original Nov. 2019)	Based on 2 Ah MLPC Carbon Anode vs high-voltage LNMO testing, compile data package of analytical findings for further development of LNMO cathode.	Re-Opened & Extended
Oct. 2021	Based on C/LNMO 2Ah MLPC testing, deliver forty 2 Ah C/LNMO MLPC containing selected electrolyte formulations to USABC for further evaluation	Reworked & On Track
Feb. 2022	Determine final electrolyte additives and formulations for 10 Ah cells	Ongoing
March 2021	Deliver thirty 10 Ah C/LNMO MLPC to USABC for further evaluation	Ongoing
Moving Target	Publication through journal paper and/or patent of any additives and/or other discoveries considered appropriate	Completed & On Going

Barriers[1]

- Cost: The cost of current high-energy lithium ion batteries is approximately 2-3x too high with raw materials being one of the main contributing factors.
- Performance: Higher energy density materials can reduce cost and weight but suffer from life and performance issues to match gas powdered vehicles' performance and customer convenience.
- Life: Next generation technologies suffer cycle, gassing and calendar life issues

[1]US DRIVE Electrochemical Energy Storage Technical Team Roadmap September 2017
https://www.energy.gov/sites/prod/files/2017/11/f39/EESTT%20roadmap%202017-10-16%20Final.pdf

Relevance

Objectives

- Analyze and understand LTO and LNMO materials through study of electrolyte interactions, gas analysis, and failure mechanisms
- To develop and evaluate LTO electrolytes and electrolyte additives that demonstrate minimal gassing, high cycle life, high power charge/discharge capabilities, wide operating temperature, and competitive cost
- To develop and evaluate LNMO electrolytes and electrolyte additives that demonstrate minimal gassing, high cycle life, high power charge/discharge capabilities, wide operating temperature, and competitive cost

Impact

- LTO is a sought-after anode material due to its improved safety, cycle and calendar life, and operating temperature range in comparison to graphite anode. By optimizing the electrolyte to mitigate the known gassing issues moves this material closer to commercial viability.
- LNMO is a high voltage (>4.9V) cathode material that increases energy density while reducing cost due to the elimination of cobalt. However, modern electrolyte is not compatible. New additives, electrolyte formulations, and mechanistic understanding can make this technology more accessible.
- Combining LNMO/LTO, which is the ultimate objective of the project, allows for a balance of the benefits of each material. The high-voltage LNMO improves the high working potential of LTO and balances the battery cost while the LTO provides improved cycle life and safety.

Approach

Multi-Layer Pouch Cell (MLPC) Performance Testing

In 2Ah and 10Ah MLPC, the following testing is being conducted to gain information on performance in tandem with gas analysis for failure mechanism understanding, surface analysis for electrolyte-electrode interaction understanding, and Manganese dissolution tracking

- -20°C Cycling
- 25°C Cycling
- 45°C Cycling
- 45°C, 1 week, 100% SOC Storage Testing
- 45°C, 4 week, 100% SOC Storage Testing

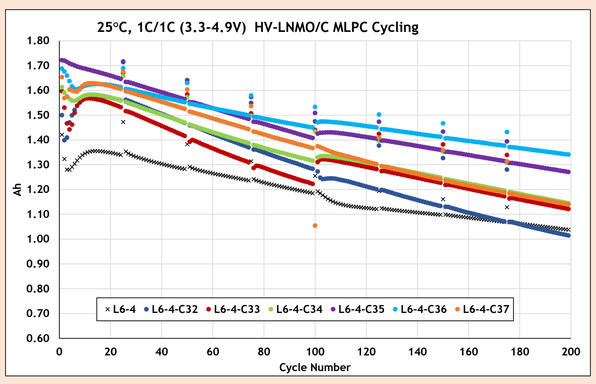
Electrolyte Intrinsic Property Study

In order to track the possible trends associated with the electrolyte properties and how they are can be applied, the following is conducted and analyzed:

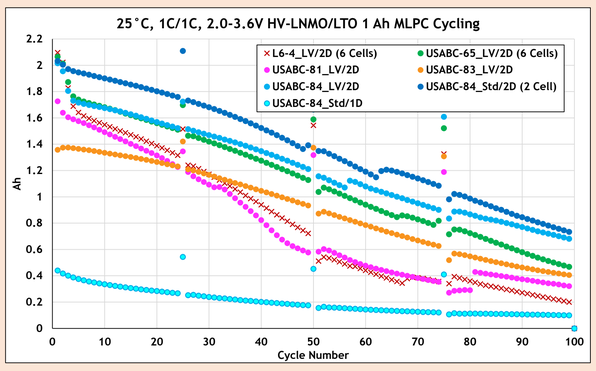
- Electrochemical Stability
  - Voltammetry & Floating Testing
- Vapor Pressure
- Flashpoint
- Viscosity (30°C and -30°C)
- Specific Conductivity (30°C and -30°C)
- Water and Hydrofluoric Acid (HF) Content
- Lithium Transference Number

Additive Synthesis

In partnership with URI, new multi-functional additives that will focus on solid electrolyte interphase (SEI) formation that improves gas generation, MN dissolution, and/or electrolyte stability are being synthesized and tested to improve the electrolyte properties and cell testing



Graphs show some of the cycling data collected for the 2Ah MLPC testing with HV-LNMO/C (top) and HV-LNMO/LTO (bottom). In HV-LNMO/C testing, additive combination C35 and C36 showed vast improvement over the L6-4 baseline. In HV-LNMO/LTO testing, 3 formulations showed improvement over the baseline.



Technical Accomplishments and Progress

Electrolyte Property Development

Target vapor pressure was reached in one formulation but did not meet conductivity targets.

Conductivity target was met in many formulations but did not align with vapor pressure targets.

Task 1: Novel Additive Development

In the past year, 2 novel compounds were synthesized (bring project total to 12 additives) with one showing improved cycling and impedance performance in pouch cell testing.

A patent has been filed for one of the twelve additives with other patent opportunity being evaluated.

Two manuscripts have been submitted relating to the novel compounds synthesized in the project.

Task 3: HV-LNMO/C MLPC Testing

This task was closed out during this year and has since been re-opened (see future work).

Investigation into the impact of particle size, morphology, age, and supplier identified a superior supplier.

Established optimized upper cutoff voltage for cell chemistry.

Additive pack combinations that improved performance at 25°C identified.

Task 4: HV-LNMO/LTO MLPC Testing

Evaluation of formation procedure led to discovery of better formation protocol to allow for more stable SEI formation.

New solvents and impact of some known solvents identified that improved the performance and impedance during 25°C cycling testing.

USABC Gap Chart of Advanced Electrolytes

Parameter		Unit	USABC Goal	End Target	Best Results	Best Single Formulations	
Cost at a yearly production volume no more than 20,000 ton/year		$/kg	< 10	8-10	12	USABC-62	USABC-20
Electrochemical Stability	Upper Voltage	V vs. Li/Li+	5	5	~ 5		
	Lower Voltage		0	0.5	1.0		
Vapor Pressure at 30°C		mm Hg	< 1	10	10	26	52
Flashpoint		°C	> 100	40	26		
Viscosity	at 30°C	cP	< 5	5	<5		
	at -30°C		< 20	20	30		
Specific Conductivity	at 30°C	mS/cm	> 12	10-12	12.66	**9.745**	**10.16**
	at -30°C		> 4	4	9.343	**1.773**	9.343
Li^+ Transference Number			> 0.35	0.3	0.2		
Components Purity		%	> 99.99	99.98	99.0-99.95		
Lithium Salt Solubility		M	1	up to 1.4	1.2	1.2	1.2
Water Content		ppm	< 20	< 15	3.2	2.41	1.24
HF Content		ppm	< 50	< 40	28.7	42.38	37.49

USABC End Target Currently Being Met
Status is above start of project but below end target
The current status is below start of project and/or end target
Equivalent to Starting Point
Measurements have not yet been conducted

USABC Gap Chart of Cell Testing

Parameter		Unit	End Target	NCM622/LTO *Retired*	HV-LNMO/LTO *Retired*	HV-LNMO/C Current
25°C Cycling (RT Cycling)	Volume Δ after 600 Cycles	mL, %	< 8%	N/A	Excessive	6.34 mL ~ 16.5%
	Capacity Retention Projected to 1500 Cycles	%	80%	N/A	58.5% After 40 Cycles	81.2% after 200 cycles
45°C Cycling (HT Cycling)	Volume Δ after 600 Cycles	mL, %	< 10%	< 10%	N/A	Excessive
	Capacity Retention Projected to 1000 Cycles	%	80%	100% after 300 Cycles	N/A	64% at 180 cycles
45°C, 1 Week at 100% SOC Storage	Retained Capacity	%	95%	95%	N/A	TBD
	Recovered Capacity	%	97%	99%	N/A	TBD
	Volume Δ	mL, %	< 10%	< 3%	N/A	TBD
45°C, 4 Week at 100% SOC Storage	Retained Capacity	%	95%	89%	N/A	TBD
	Recovered Capacity	%	97%	99%	N/A	TBD
	Volume Δ	mL, %	< 10%	< 3%	N/A	TBD

USABC End Target Currently Being Met
Status is above start of project but below end target
The current status is below start of project and/or end target
Equivalent to Start of Project
Not yet measured

Remaining Challenges and Barriers

- HV-LNMO cathode stability, quality, and optimization is an ongoing challenge that our analytical investigations keep working to overcome while not causing disruptions to the project.
- Chemical property testing of the electrolyte formulations are showing that accomplishing targets all at once are challenging. Meeting a single target is not but aligning all the desired chemical properties is a challenge. In addition, meeting these targets does not guarantee good performance in a real cell adding another layer to this challenge.
- While impedance and cycling performance has been able to be improved, the impact to gas generation and low first cycle efficiency is still a challenge that needs to be overcome in order to make this project and material successful.

Proposed Future Research

- Based on discussions within the research group, data being collected, and targets for the project the future research has been redesigned to focus on HV-LNMO with carbon anode development in place of HV-LNMO with LTO anode development.
- 2 Ah MLPC and 10 Ah MLPC testing at 25°C and 45°C will still be conducted as described in the approach section of this poster.
- Chemical property testing will still be conducted as described in the approach section of this poster.
- While this is an electrolyte project focusing on formulation and additive development, it is encouraged to continue HV-LNMO cathode material analysis. This will be carried out time permitting throughout the remainder of this project.

Summary

This project underwent a renovation given the circumstances of last year. The accomplishments include paper and patent submission, novel additive development, new solvent identification, and cathode material discovery. The continued impact of this project is to allow for low cost, high energy density materials to become commercially viable through the development of high voltage electrolyte in support of the high voltage cathode material.

This presentation does not contain any proprietary; confidential, or otherwise restricted information.