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Towards Solventless Processing of Thick Electron-Beam (EB) Cured Lithium-Ion Battery Cathodes

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Oak Ridge National Laboratory

FY21 VTO Annual Merit Review

June 23, 2021

Project ID: BAT475

U.S. DEPARTMENT OF ENERGY

Overview

Timeline

- Task Start: 10/1/14
- Task End: 9/30/22
- Percent Complete: 85%

Budget

- $300k in FY20
- $300k in FY21

Barriers

- Barriers addressed
  - By 2022, further reduce EV battery cost to $80-100/kWh.
  - Materials processing cost reduction of at least 50% and electrode thickness increase of ≥2×.
  - Achieve deep discharge cycling target of 1000 cycles for EVs (2022) at high power density.

Partners

- Interactions/Collaborations
  - Equipment Suppliers: PCT Ebeam and Integration, Keyland Polymer.
  - Battery Manufacturers: XALT Energy, Navitas Systems
  - Materials Suppliers: BASF, Toda America, Allnex, Keyland Polymer.
- Project Lead: ORNL

Objectives & Relevance

- **<u>Main Objective</u>: To achieve 1) significant electrode process energy savings; 2) ultra-high electrode processing speed; and 3) utilize much more compact equipment than conventional drying ovens.**
  - EB treatment is a fast, robust materials processing technology.
  - Low cost and excellent compatibility with high-volume materials production.
  - Unmatched throughput: ≥600 m^2/min throughput can be achieved based on ≥300 m/min line speed for roll widths up to 2 m ($1.5-2.0M installed with footprint ~10 m^2).
  - Thicker electrodes: It is expected that cathode coatings of **several hundred microns** can be processed at ~150 m/min or with a larger equipment footprint.
  - Excellent energy efficiency – Electrical efficiencies ≥60% are possible.
  - Environmentally friendly – EB processing requires no solvent and no photoinitiator and has low emissions.

- **Relevance to Barriers and Targets**
  - Significantly enabling technology for achieving ultimate EV battery pack cost of $80/kWh through substantial materials processing cost reduction.
  - *Further enables cell energy density improvement through electrode thickness increases of at least 2× **<u>and</u>** is an enabling technology for solid-state battery cathodes and electrolytes.*
  - *Develops deposition methods for electrode manufacturing requiring little or no solvent.*

Milestones

Installation and implementation of roll-to-roll electron beam curing line at ORNL.

Completed on 9/30/2020 ✓

Fabrication of cathode/electrolyte bilayer with high areal capacity via EB curing.

- Coat a 30 mg/cm^2 NMC 811 cathode and a 25-micron PEO/LLZO composite solid electrolyte on newly commissioned ebeam Technologies R2R line at 30 ft/min.

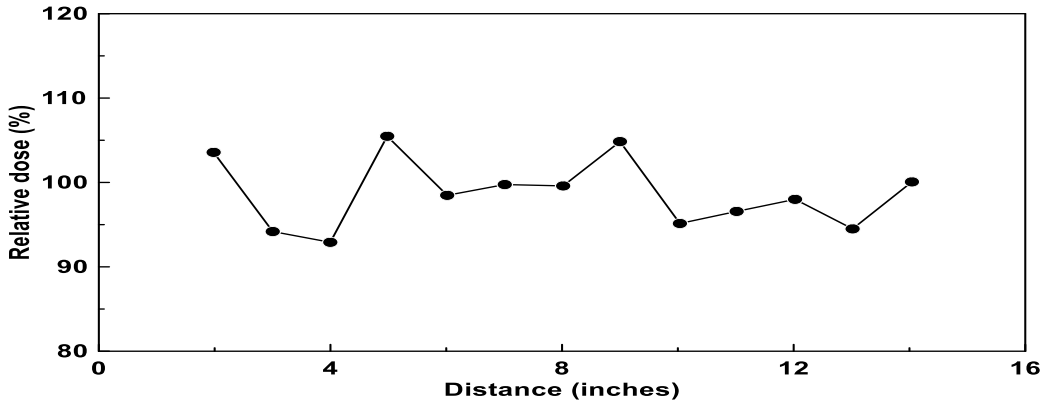
In progress, 6/30/2021

Approach

- **Major problems to be addressed:**
  - Conventional solvent drying process limits the coating speed and requires long drying lines to accommodate high coating speed.
  - These drying lines are operating and capital expense intensive and require a large amount of battery plant space.
  - Cost of organic solvents and solvent handling are prohibitive in terms of processing cost and capital expense.
- **Overall technical approach and strategy:**
  1. Phase 1 – Demonstrate the technology's key differentiating attributes of high throughput and thick layer processing (FY15-16).
  2. Phase 2 – Address the key challenges of EB curing parameters and resulting material performance; develop thick coating methods requiring little or no solvent. (FY17-18).
  3. Phase 3 – Demonstrate thick cathode coatings and optimized curing system in conjunction with a high-speed coating line together with a key equipment partner and battery manufacturer (FY19-20).
  4. Phase 4 – Installation, commissioning and operation of a custom roll-to-roll EB curing line at BMF and demonstration of solid-state battery cathode and electrolyte production (FY20-21).

Design, Purchasing, Installation, and Commissioning of R2R EB Curing Pilot Line at ORNL BMF Was Completed During Phase 4

EB pilot line was successfully installed and in full operating condition at the ORNL BMF.



Dose uniformity across the dosimeter strip.

	Key parameters
Voltage	120-300 keV
Width	15 inches
Line speed	3-30 feet per minute
Dose	750 kGy- m/min at 300 kV
Inert	self-shielded and N_2 inerted ≤ 200 ppm of oxygen

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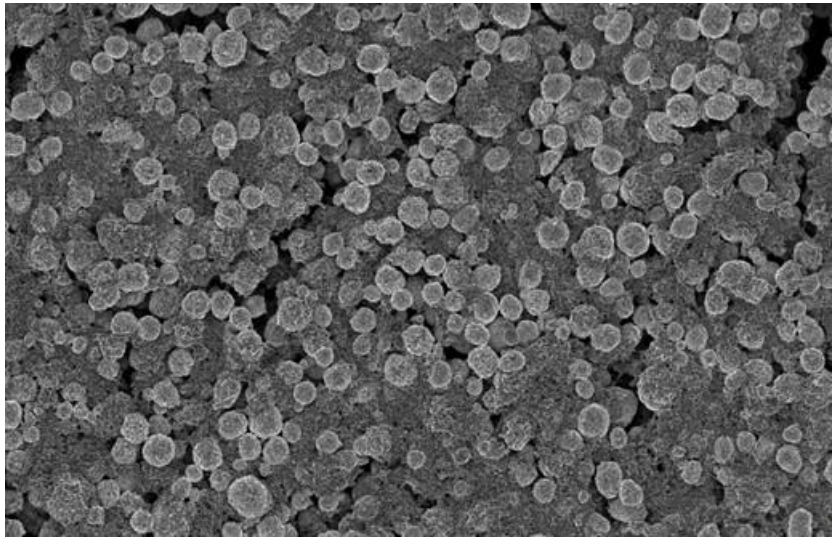
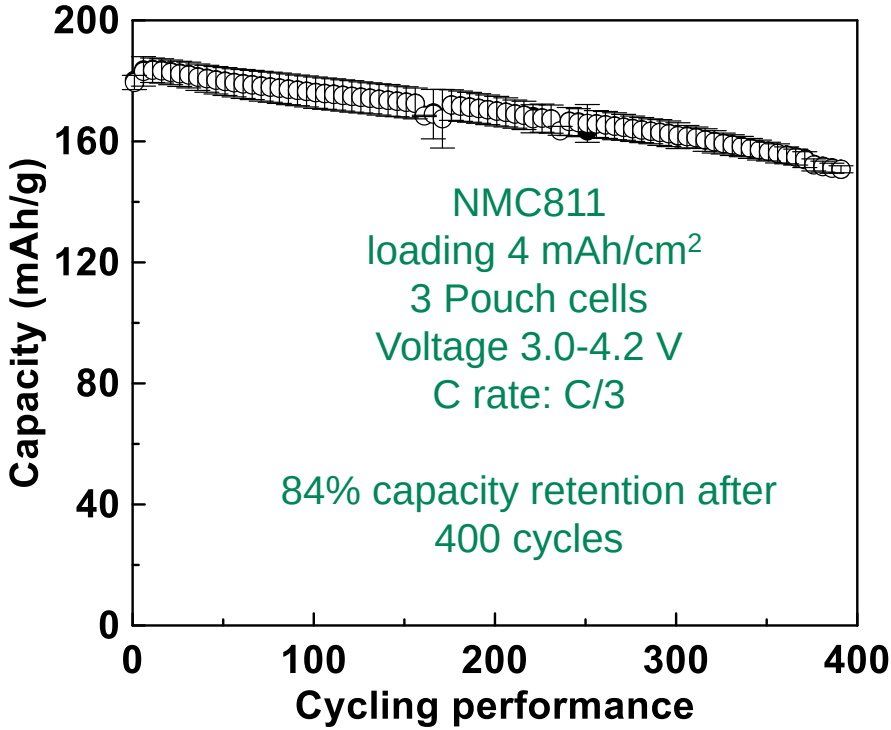
Thick Cathodes Processed by EB Curing Exhibited Excellent Capacity Retention of 84% Through 400 USABC Cycles

Electron beam curing of thick NMC811 cathode

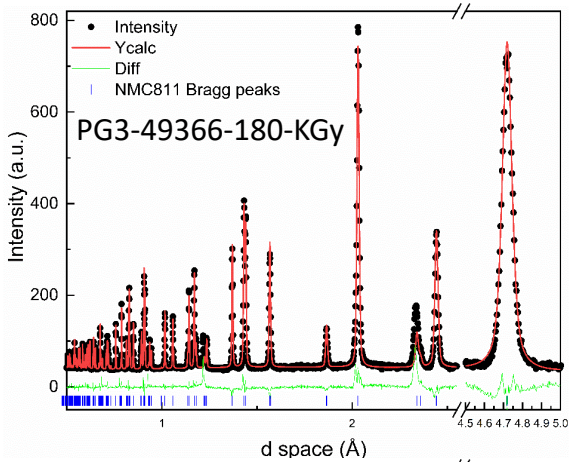


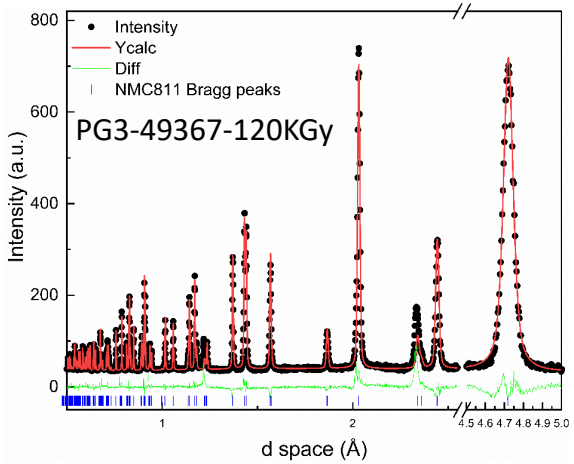
Electrode morphology shows typical NMC particles within carbon black network

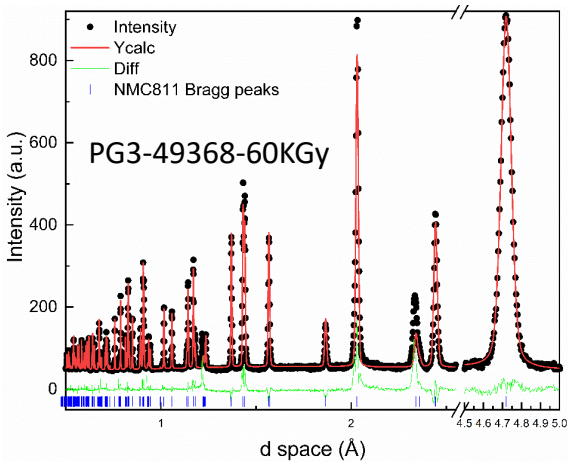


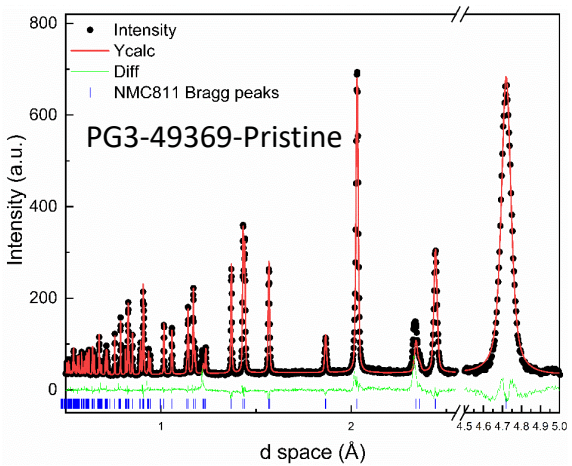
Thick Cathodes Processed by EB Curing Exhibited Excellent Capacity Retention of 84% Through 400 USABC Cycles

EB Radiation Has Little Effect on the Cathode Active Material Crystallinity



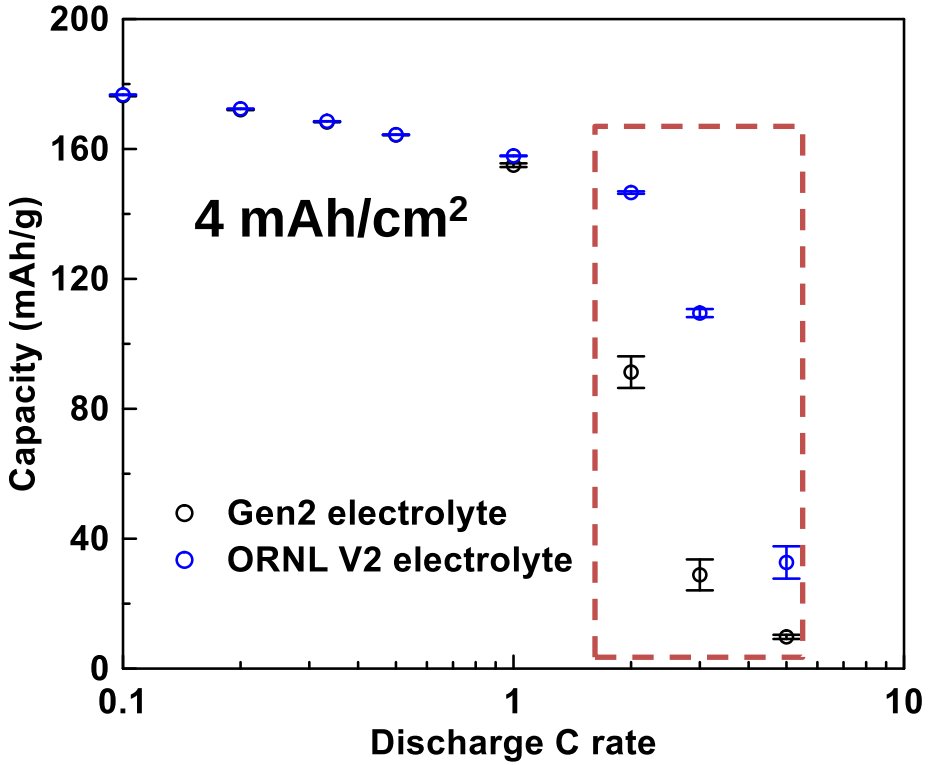


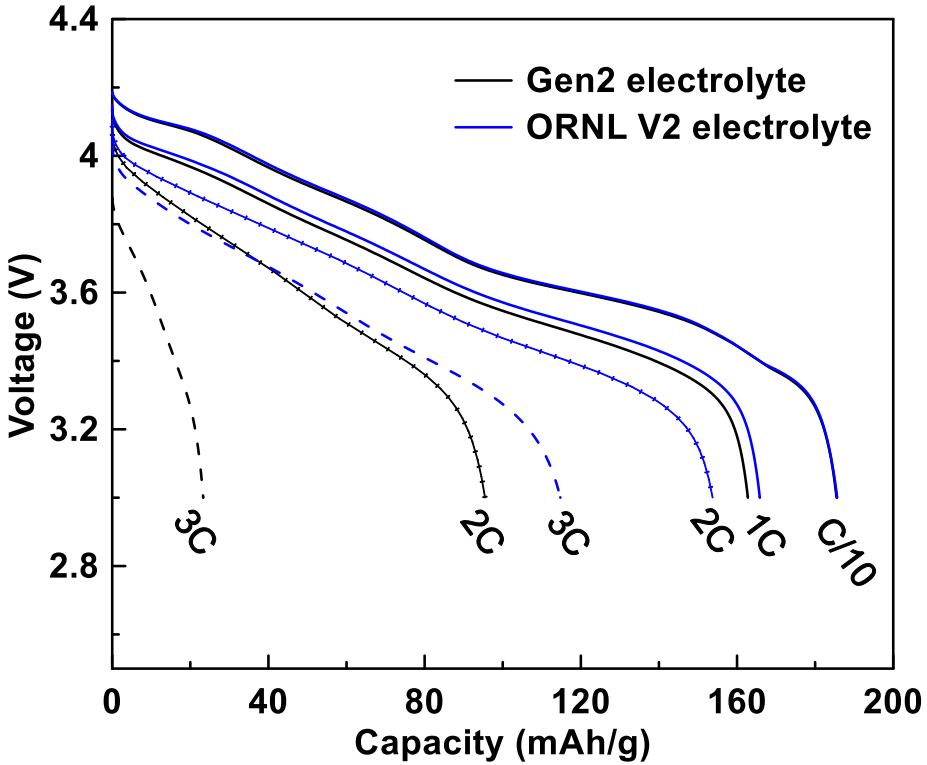




Name	Radiation dose kGy	a	c	z	Cation Mixing
NMC811	180	2.86368	14.15683	0.24075	0.01199
	120	2.86334	14.15814	0.24078	0.01317
	60	2.86325	14.1544	0.24094	0.03506
	N/A	2.86341	14.15585	0.24072	0.00859
NMC622	180	2.85978	14.1671	0.24095	0.04516
	120	2.86078	14.17011	0.24056	0.00673
	60	2.8609	14.17033	0.24074	0.01653
	N/A	2.86027	14.16857	0.24079	0.02356

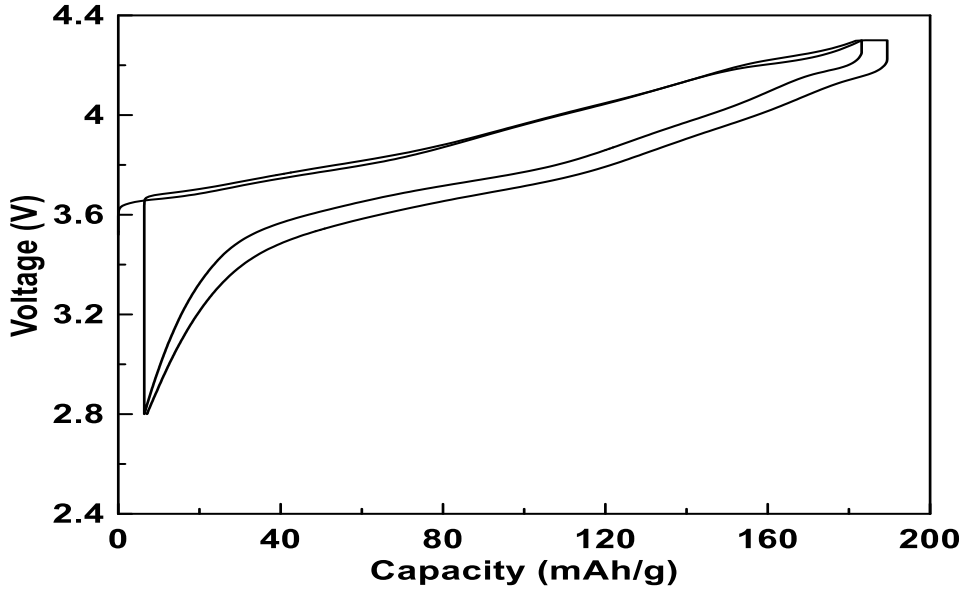
EB Curing of Thick Cathodes Combined with ORNL Extreme Fast Charging Electrolyte Significantly Boosts High-Power Performance

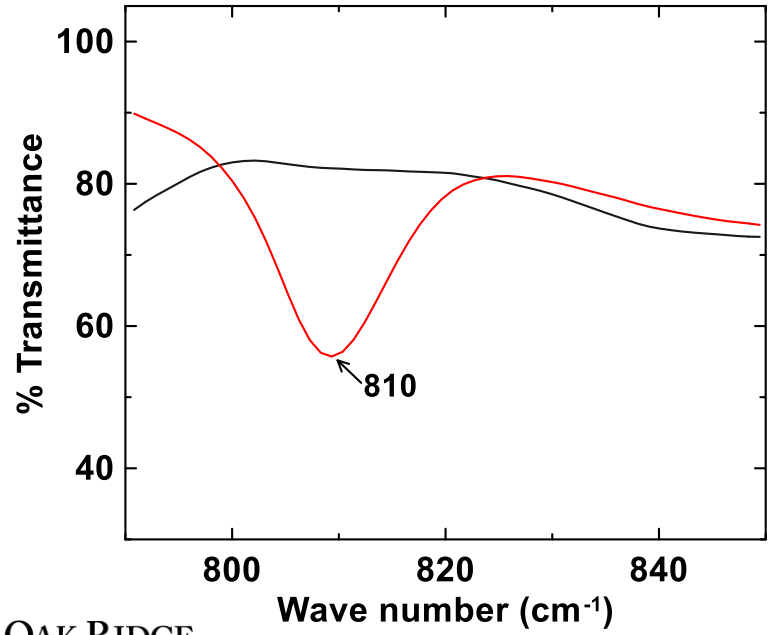




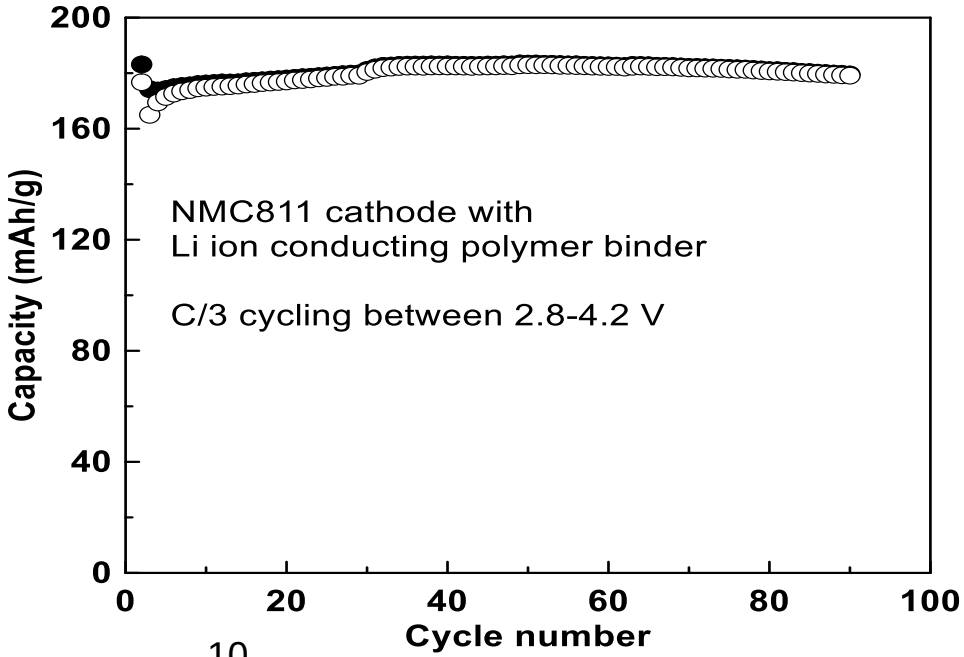
Rate/power performance at 2C and 3C is highly improved with optimized electrolyte formulation.

Exploring Li^+ Conducting Polymer as a Binder for Composite Cathode





FTIR confirmed full cross-linking during EB curing



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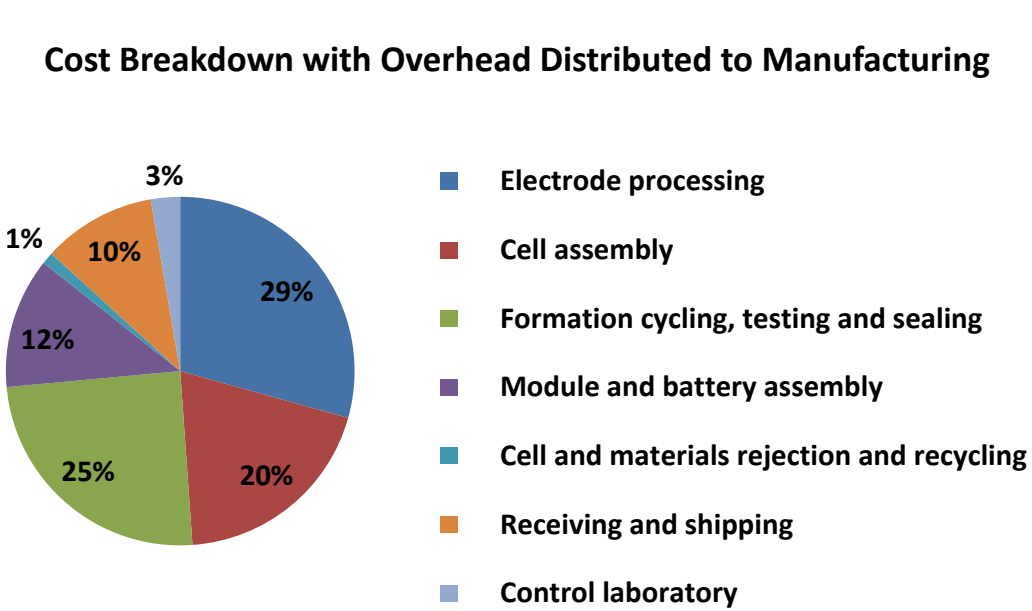
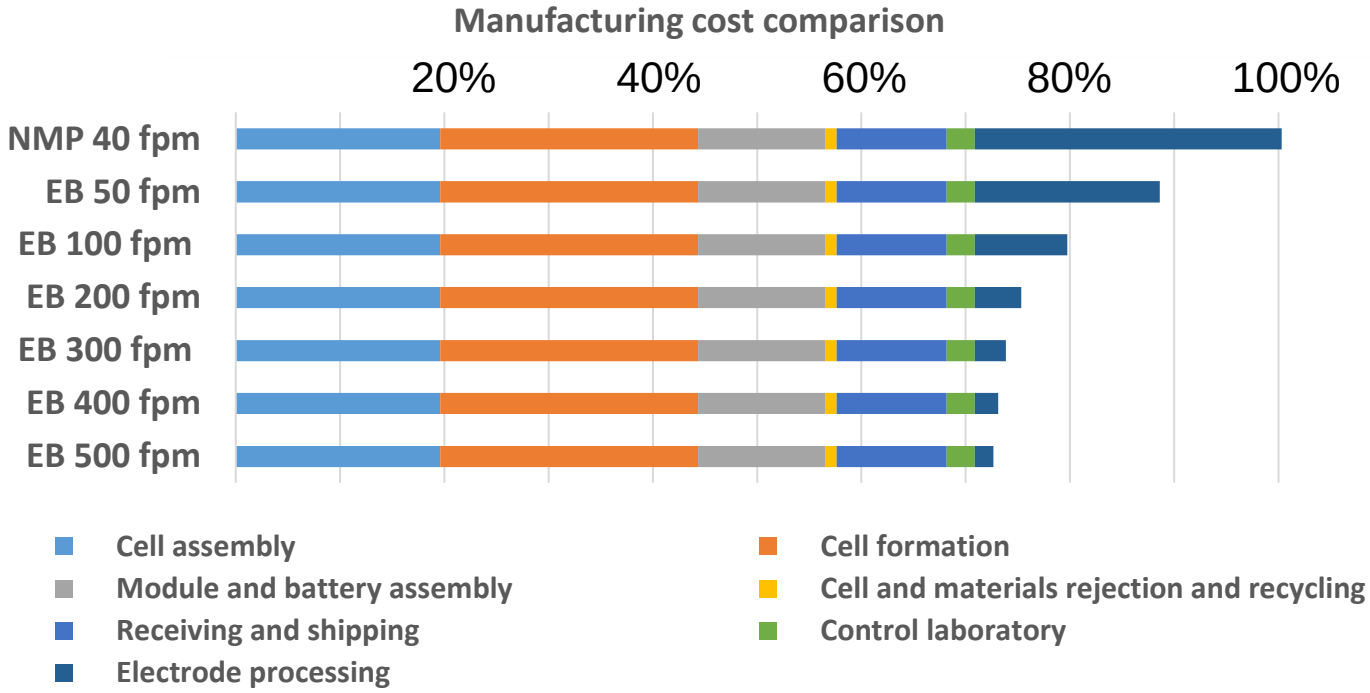
Substantial Operating and Capital Cost Reduction Can Be Realized with EB Processing



Cost breakdown of the manufacturing when NMP drying and recovery are used in electrode processing.



Electrode processing cost reduction by using EB processing compared to NMP drying/recovering process.

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Collaborations

- Partners
  - Equipment Suppliers: PCT Ebeam and Integration, Keyland Polymer, B&W MEGTEC, Eastman Kodak
  - Battery Manufacturers: XALT Energy, Navitas Systems
  - Raw Materials Suppliers: BASF, Allnex, Keyland Polymer, Superior Graphite, Denka

Selected Responses to Specific FY20 DOE AMR Reviewer Comments

Not reviewed in FY20.

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Proposed Future Research (FY21-22)

1. Electron beam curing of matching anodes

   Demonstrate no more than 20% capacity fade through 500 cycles at 0.33C/-0.33C in pouch cells with both cathode and anode EB cured with the ORNL roll-to-roll EB curing line.

2. EB/UV R2R polymer electrolyte and composite cathode manufacturing

   Demonstrate cycling performance and high energy density of solid-state Li cell using EB/UV cured cathode and EB/UV cured polymer composite electrolyte.

3. Understanding of processing-microstructure-performance relationships

   Characterizing electrode microstructures after different EB processing parameters, testing cells performance, and stablishing the connections in processing-microstructure-performance relationship.

Any proposed future work is subject to change based on funding levels.

Summary

- Design, Purchasing, Installation, and Commissioning of R2R EB Curing Pilot Line at ORNL BMF Was Completed During Phase 4.
- Thick Cathodes Processed by EB Curing Exhibited Excellent Capacity Retention of 84% Through 400 USABC Cycles.
- EB Radiation Has Little Effect on the Cathode Active Material Crystallinity in EB curing process.
- Li ion Conducting Polymer has been demonstrated as a Binder for Composite Cathode with stable cycling.
- Power /rate performance of EB Curing of Thick Cathodes can be significantly improved by state-of-the-art electrolyte formulation.
- Business analysis shows Substantial Operating and Capital Cost Reduction Can Be Realized with EB Processing.
- Commercialization: High likelihood of technology transfer because of strong industrial collaboration, significant electrode production cost reduction, and impact on cell energy density.

Acknowledgements

- U.S. DOE Office of Energy Efficiency and Renewable Energy (EERE) Vehicle Technologies Office (Director: David Howell and Program Manager: Peter Faguy)
- ORNL Contributors:
  - Ilias Belharouak
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  - Andrew Todd

Technical Collaborators

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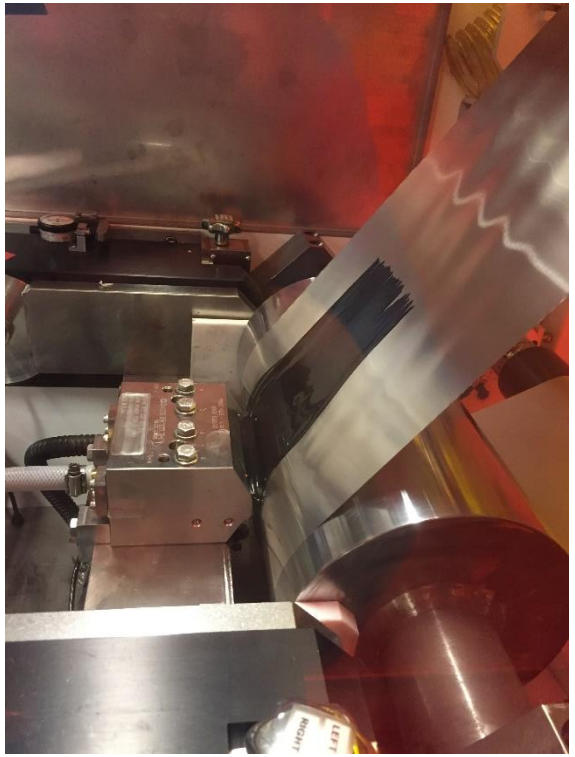
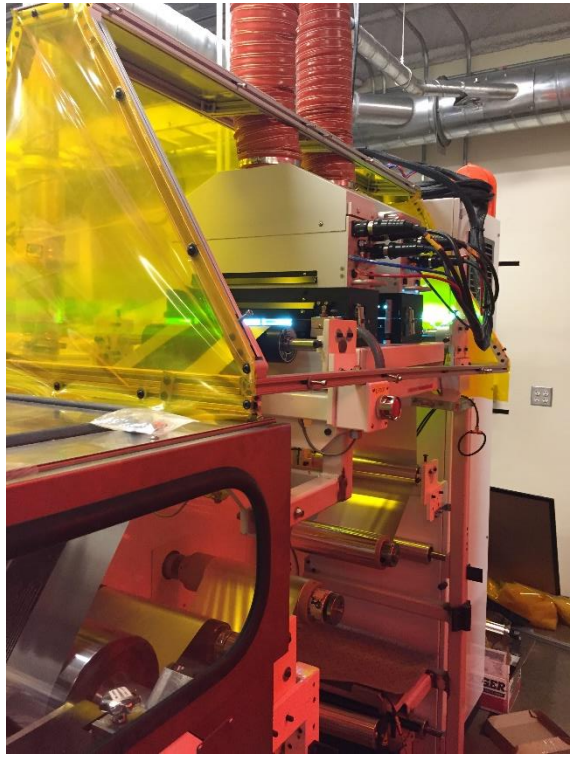
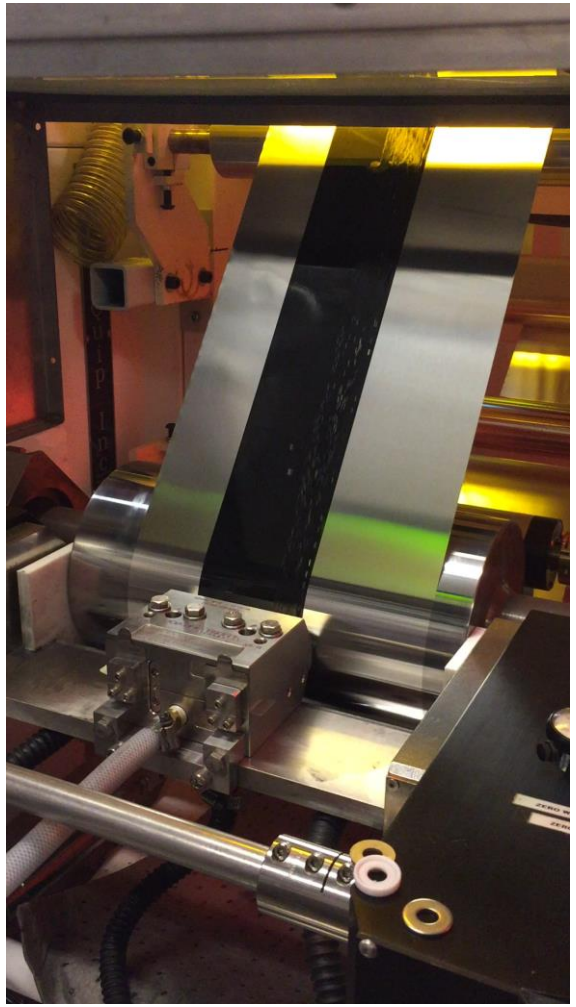
- Kevin Otto
- Karl E. Swanson
- Sage Schissel
- Michael Bielmann
- Dave Ventola
- Kevin Dahlberg
- Mike Wixom
- Dan Occorr

Backup slides

Commissioning of R2R UV Curing Line at ORNL

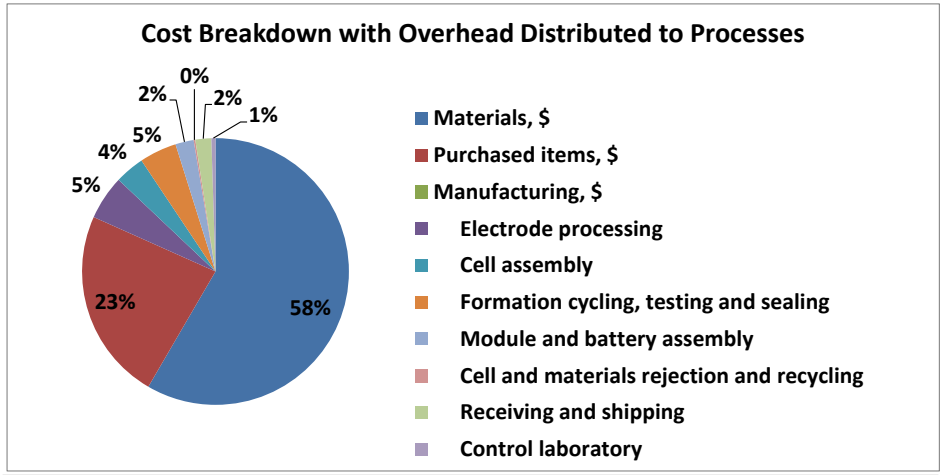


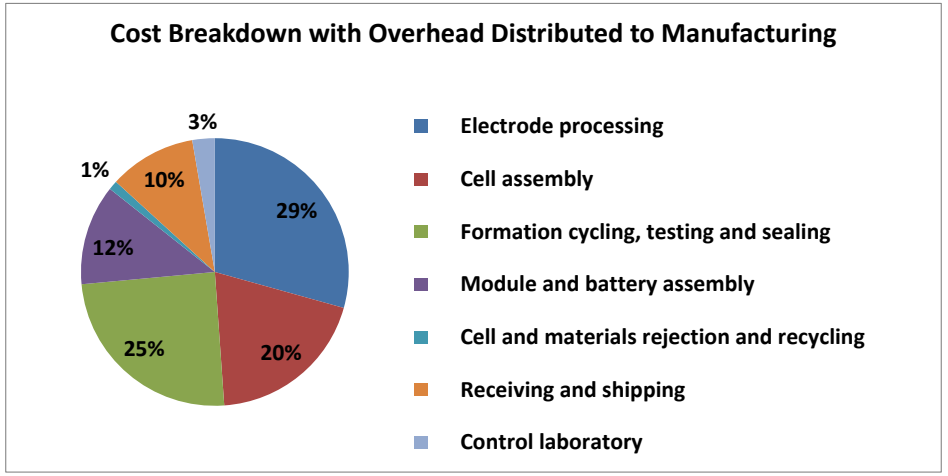




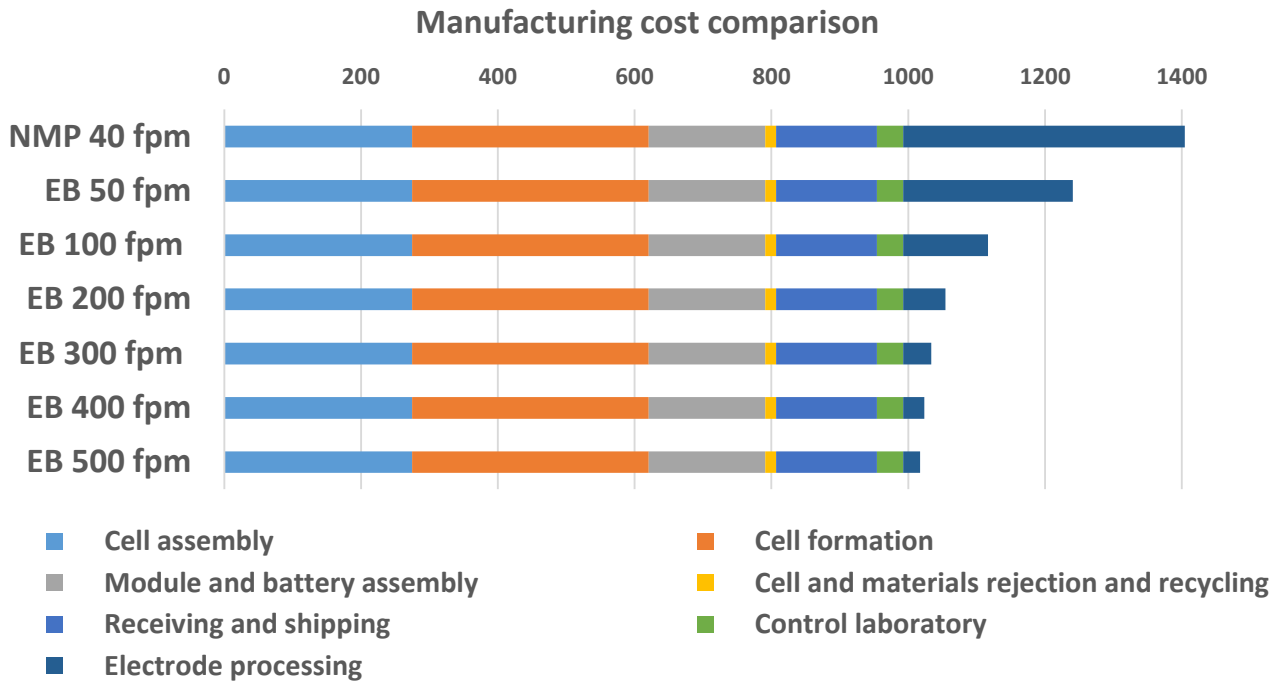
- UV curing advantages: cost of equipment low
- Suitable for thin or transparent coatings
- Not suitable for thick/opaque coatings

Substantial Operating and Capital Cost Reduction Can Be Realized with EB Processing





Top: Cost breakdown distributed to processes; bottom: cost breakdown of the manufacturing when NMP drying and recovery are used in electrode processing.



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