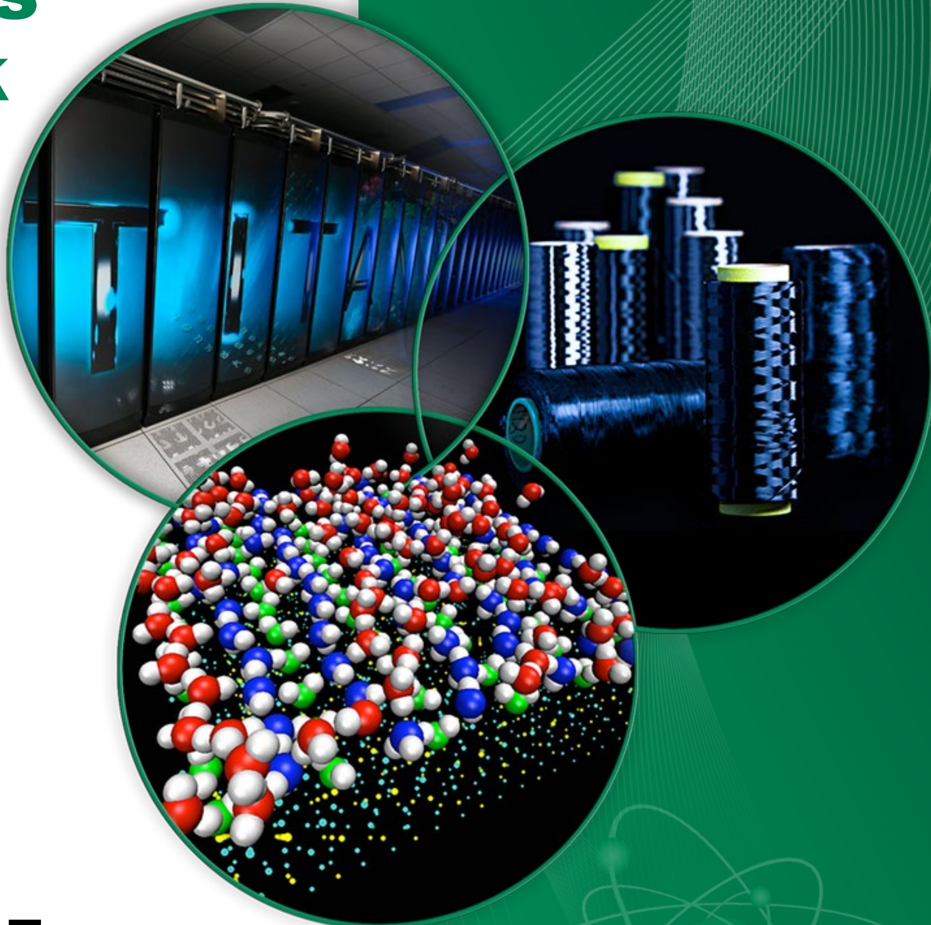
Towards Solventless Processing of Thick Electron-Beam (EB) Cured Lithium-Ion Battery Cathodes



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bat207

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Overview

Timeline

- Task Start: 10/1/14
- Task End: 9/30/21
- Percent Complete: 70%

Barriers

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

Budget

- $350k in FY18
- $400k in FY19
- $400k in FY20 (planned)

Partners

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

Objectives & Relevance

- **Main Objective: 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 $80-100/kWh through substantial materials processing cost reduction.
  - *Further enables cell energy density improvement through electrode thickness increases of at least 2×.*
  - *Develops deposition methods for electrode manufacturing requiring little or no solvent.*

Task Milestones and Progress

<u>12/31/18 (Quarterly Progress Measure): Combined Electrostatic Spraying and Electron Beam (EB) Curing Demonstration</u>

Demonstrate no more than 20% capacity fade through 300 cycles at 0.33C/-0.33C in single unit pouch cells with electrode processed through combination of solvent-free electrostatic spraying and EB curing methods for NMC811 composite electrode manufacturing with industrial partners and Keyland Polymer at 25 mg/cm^2 loading.

- ✓ Delayed to 6/30/2019 due to equipment issue

<u>9/30/19 (Annual Milestone Stretch): Thick electron beam (EB) cured cathode processing demonstration at ultra-high line speeds.</u>

Demonstrate no more than 20% capacity fade through 500 cycles at 0.33C/-0.33C in 1.5 Ah pouch cells with optimized cathode EB curing formulation and NMC 811 areal loading of 30 mg/cm^2 (structured and unstructured coatings) at a curing speed of 150-200 m/min.

- ✓ On track

<u>Go/No-Go (Cathode EB Curing Speed and Areal Loading, 9/30/19): Demonstrate 30 mg/cm2 NMC811 cathode coating areal weight with full EB cure and down-selected electrode formulation with PCT</u> at 150-200 m/min.

If this outcome is a no-go, then either the EB formulation will be redesigned, the industrial partner production process will be modified (nitrogen blanket, etc.), or both for FY20.

- ✓ On track

Approach

- **Major problems to be addressed:**
  - Conventional solvent primary drying ovens for lithium-ion electrodes are not compatible with high line speeds or must include long drying lines to accommodate high line 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 ultra-thick (38 mg/cm^2 NMC 622) coating methods requiring little or no solvent. (FY17-18).
  3. Phase 3 – Demonstrate ultra-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 (FY20-21)

Technical Accomplishments – Executive Summary

- FY18Q3:
  - ✓ Completed 1000 cycles of 500 ft/min EB curing of NMC 532 cathode with 25 mg/cm^2 loading.
- FY18Q4:
  - ✓ High speed EB curing of thick NMC622, NMC811 cathode at PCT.
  - ✓ Cell performance evaluation of the obtained electrodes.
- FY19Q1:
  - ✓ Collaboration with Keyland Polymer on new resin development and Li-ion cell development.
  - ✓ Electrostatic spraying trials on NMC811 with Keyland Polymer.
- FY19Q2:
  - ✓ Working with US equipment manufacturer on purchasing a roll-to-roll EB curing pilot line at ORNL.
  - ✓ Evaluation of low Tg resins on Ni-rich (low cobalt) cathode materials.

High speed EB curing of NMC622 and NMC811 electrodes shows successful results

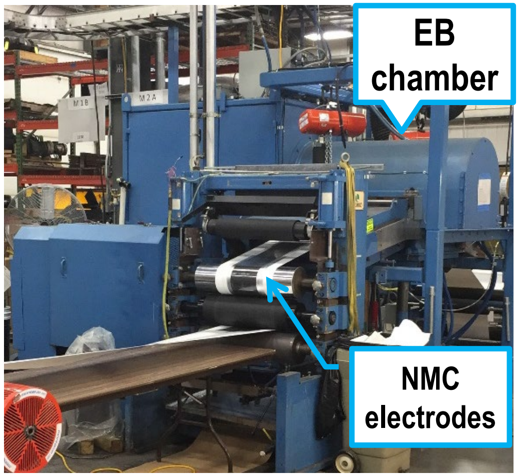


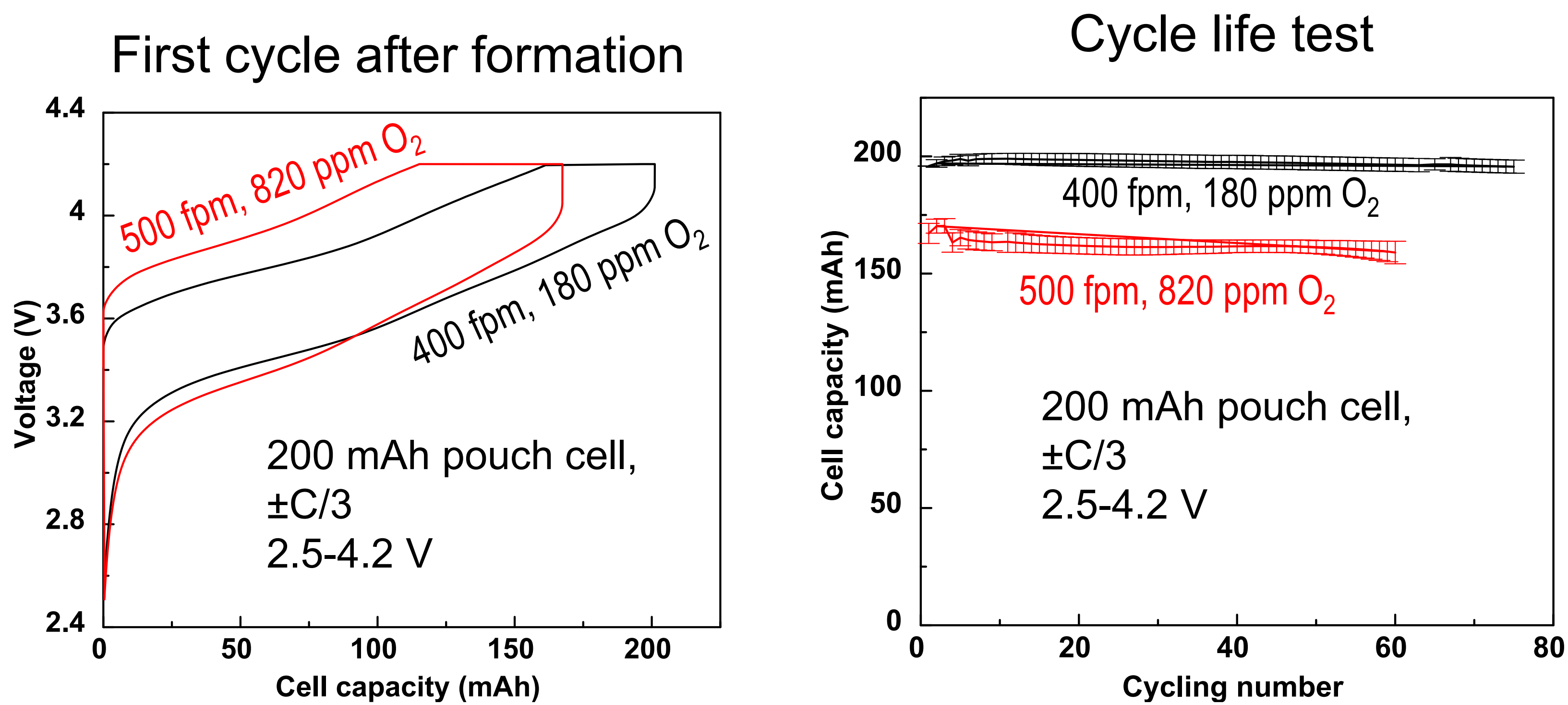
Table 1. Summary of the high speed curing experimental runs at PCT.

Run No.	Sample Name	Experimental details	EB conditions	Oxygen level
I	NMC622 - 30 mg/cm^2 NMC811 - 30 mg/cm^2	covered and N_2 inerted using Stretch-tite film.	290kV/60kGy 500 fpm	820 ppm
II	NMC622 - 30 mg/cm^2 NMC811 - 30 mg/cm^2	covered and N_2 inerted using Stretch-tite film.	290kV/75kGy 400 fpm	180 ppm

Table 2. Summary of the high speed curing (A-dosimeter under NMC622 coating, B-dosimeter under NMC811 coating)

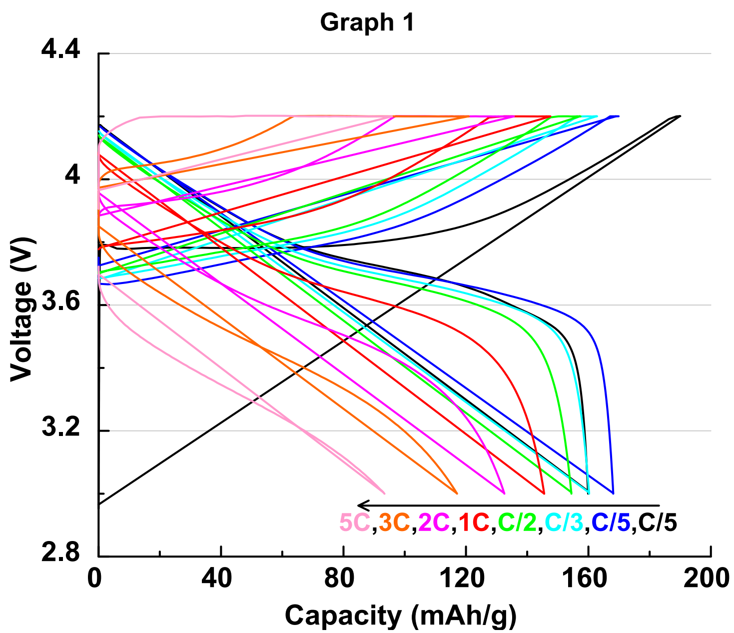
Test #I: 60 kGy / 290 kV / 500 fpm / 300gsm									
	Dose (kGy)		Dose (kGy)	*Estimated*	Dose (kGy)		Dose (kGy)	*Estimated*	Dose (kGy)
Top L	68.3	A1 Bottom	55.3	A1 Bottom No Al	68	B1 Bottom	51.2	B1 Bottom No Al	68
Top C	69.2	A2 Bottom	53.2	A2 Bottom No Al	69	B2 Bottom	50.3	B2 Bottom No Al	69
Top R	66.8	A3 Bottom	56.6	A3 Bottom No Al	67	B3 Bottom	47.8	B3 Bottom No Al	67
Test #II: 75 kGy / 290 kV / 400 fpm / 300gsm									
	Dose (kGy)		Dose (kGy)	*Estimated*	Dose (kGy)		Dose (kGy)	*Estimated*	Dose (kGy)
Top L	Over Range	A1 Bottom	55.9	A1 Bottom No Al	80	B1 Bottom	57.5	B1 Bottom No Al	80
Top C	Over Range	A2 Bottom	53.4	A2 Bottom No Al	80	B2 Bottom	57.5	B2 Bottom No Al	80
Top R	78.3	A3 Bottom	53.9	A3 Bottom No Al	78	B3 Bottom	56.1	B3 Bottom No Al	78

N_2 Blanket for O_2 Reduction Is Critical for Full Crosslinking (5 mAh/cm^2, NMC811 Results)

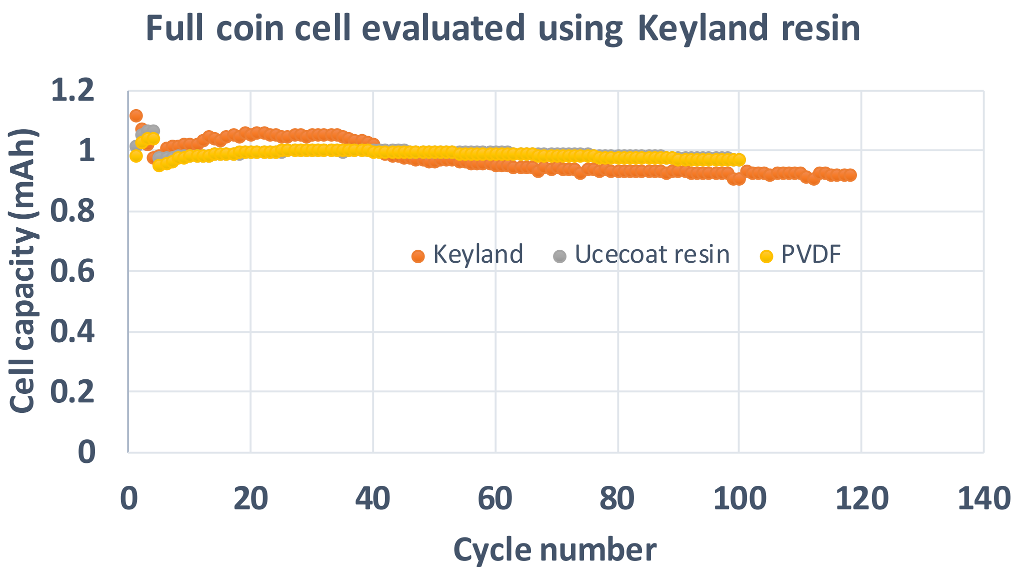


- 200 mAh pouch cells with two NMC811 electrodes fabricated with different EB curing conditions
- Higher capacity and lower polarization in electrodes processed at lower O_2 level

Development of New EB Curable Resin for NMC622 cathode with Keyland Polymer



Voltage-capacity curves of EB cured NMC cathodes at different rates.



Cycling performance of full coin cells with NMC cathodes using different binders.

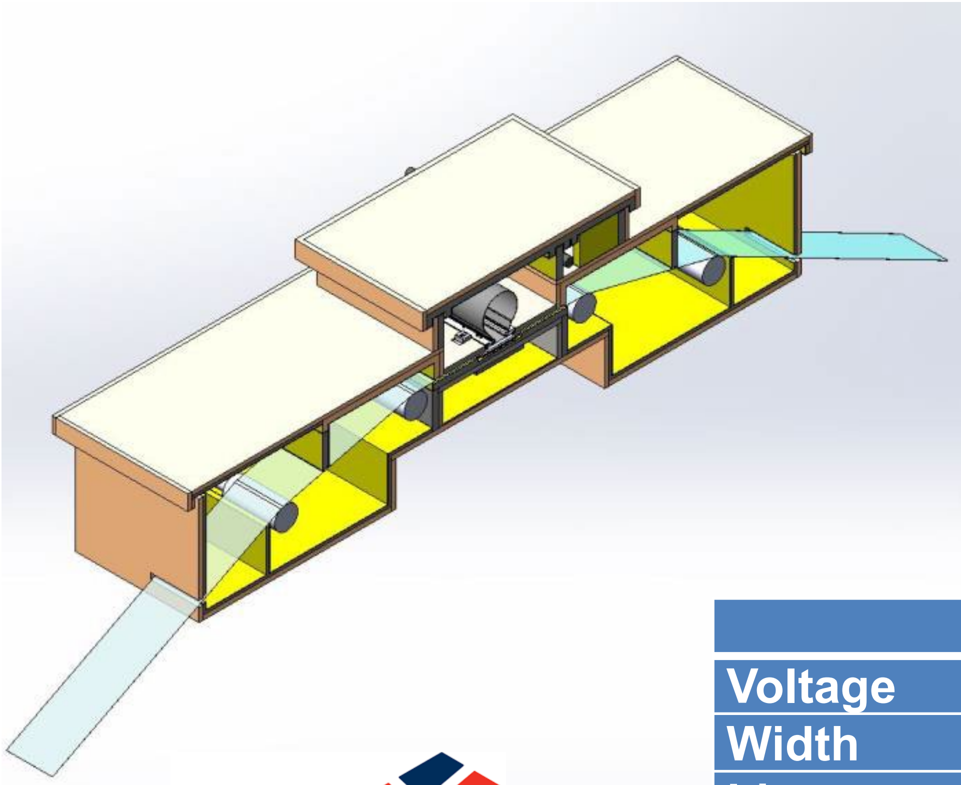
- Collaboration with Keyland polymer on new resin development for smaller colloidal size.
- Full coin cell demonstrated equally good performance

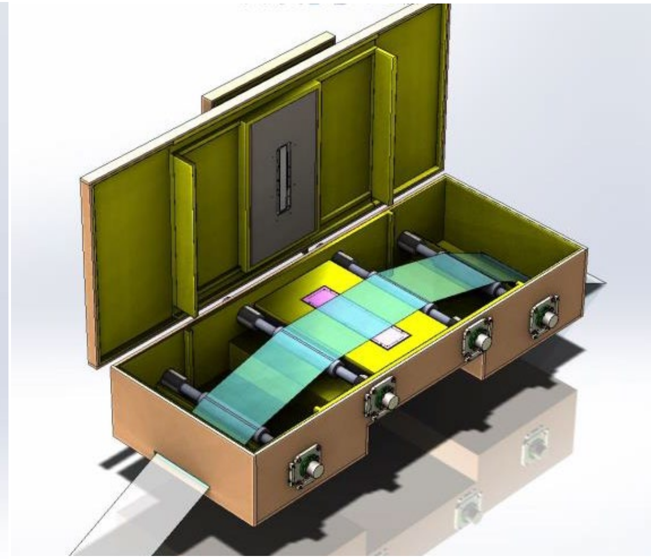
New Processing Protocols Were Developed for Electrostatic Spraying

Key processing parameters were identified for electrostatic spraying to have good adhesion and quality

- Collaboration with Keyland polymer on electrostatic spraying
- Key processing protocols were identified for better adhesion and quality
- Plan to study intermittent spraying for higher areal loading

New Capital Investment in Pilot Scale Roll-to-Roll EB Curing Equipment at ORNL





PCT

	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

Courtesy: PCT Ebeam and Integration

Collaborations

- Partners
  - Equipment Suppliers: PCT, Keyland Polymer, B&W MEGTEC, Eastman Kodak
  - Battery Manufacturers: XALT Energy, Navitas Systems
  - Raw Materials Suppliers: TODA America, Keyland Polymer, Superior graphite, Denka

- Collaborative Activities
  - Extensive EB trials were completed at Keyland Polymers, NEO Beam and PCT ebeam and integration in from 2015 to 2019.
  - High speed curing at 500 fpm has been demonstrated at PCT pilot coating and curing line in Davenport, IA.
  - Lab-scale "power" coatings are under evaluation at Keyland Polymer.

Selected Responses to Specific FY18 DOE AMR Reviewer Comments

Project not reviewed in FY18.

Proposed Future Research (FY19-20)

- Assemble pouch cells with EB cured electrodes and evaluate their electrochemical performance
- Installation of roll-to-roll EB curing pilot line at ORNL.
- Adjustment and Optimization of the EB curing Pilot line at ORNL.
- Demonstrate EB cured Thick NMC811 electrode (30 mg/cm^2, 5 mAh/cm^2) using ORNL EB curing line.
- Resin development for flexibility of the EB cured coating.
- Resin development for electrostatic spraying with industrial partner.

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

Summary

- **Objective:** To achieve 1) significant process energy savings; 2) ultra-high electrode processing speed; and 3) utilize much more compact production equipment.
- **Approach:** Three-phase approach from formulation chemistry to full-scale production.
  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 coating methods requiring little or no solvent. (FY17-18).
  3. Phase 3 – Demonstrate an optimized curing system 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 (FY20-21)
- **Technical:** ***500 feet/min EB curing pilot line demonstration, Pouch cell performance evaluation, dry powder electrostatic spraying with key industrial partner.***
- **Collaborators:** High-speed EB curing scale-up at the PCT pilot line in Davenport, IA. Electrostatic spraying powder coating evaluation at Keyland Polymer in Cleveland, OH. Plans to investigate other high-speed coating technologies with high solids (low solvent) content with either B&W MEGTEC or Eastman Kodak.
- **Commercialization:** High likelihood of technology transfer because of strong industrial collaboration, significant electrode production cost reduction, and impact on cell energy density.

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- Kevin Dahlberg
- Lisa Stevenson
- Mike Wixom
- Dan Occorr

Information Dissemination and Commercialization

Patent applications

- A Method of Solvent-Free Manufacturing of Composite Electrodes Incorporating Radiation Curable Binders, Z. Du, C. J. Janke, J. Li, D. L. Wood, III, C. Daniel, Appl. No.: 15/966,840.
- Manufacturing of Thick Composite Electrode Using Solvent Mixtures, Z. Du, J. Li, D. L. Wood, III, C. Daniel, Appl. No.: 15/965,242

Journal Papers and Presentations

- Z. Du, C.J. Janke, J. Li, and D. L. Wood III, “High−Speed Electron Beam Curing of Thick Electrode for High Energy Density Li-ion Batteries”, *Green Energy & Environment, in press*.
- Z. Du, C.J. Janke, J. Li, and D. L. Wood III, Radiation Curing and Its Application in Li Cells, 235th ECS meeting, Dallas, TX.

Thank you for your attention!