

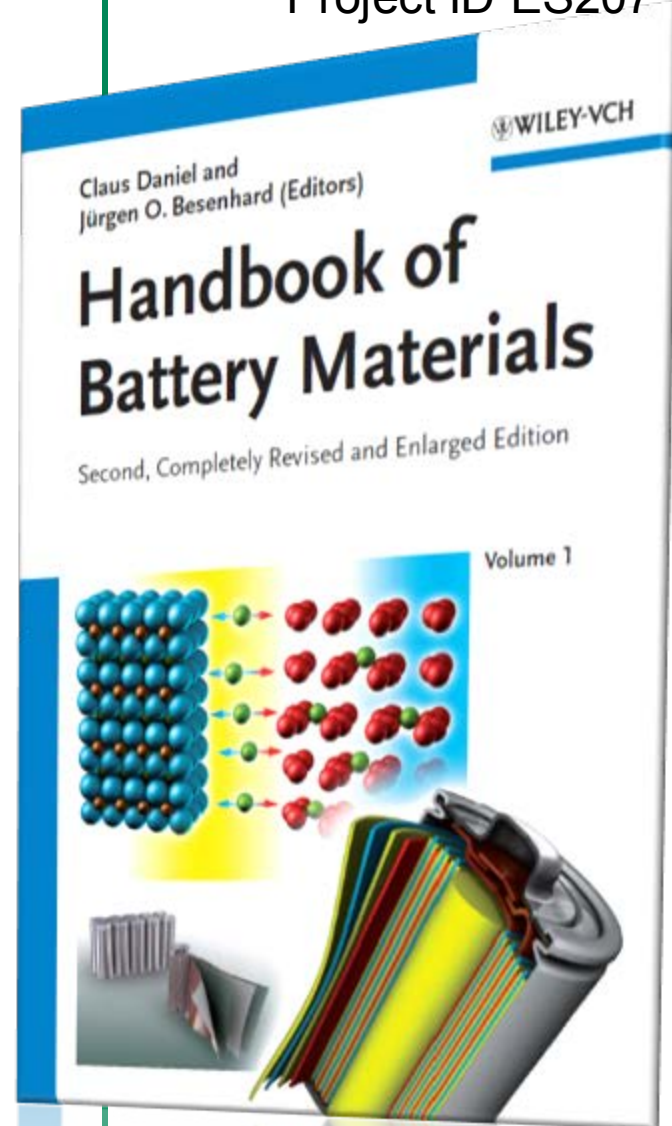
Manufacturability Study and Scale- Up for Large Format Lithium Ion Batteries

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Oak Ridge National Laboratory

June 18, 2014

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U.S. DEPARTMENT OF
ENERGY

Energy Efficiency &
Renewable Energy



OAK RIDGE NATIONAL LABORATORY

MANAGED BY UT-BATTELLE FOR THE DEPARTMENT OF ENERGY

Overview

Timeline

- Project Start: 10/1/12
- Project End: 9/30/16
- Percent Complete: 41%

Budget

- Total project funding
 - \$1,320K
- Anticipated \$330k in FY14

Barriers

- Barriers Addressed
 - By 2020, reduce EV battery cost to \$125/kWh.
 - Demonstrate scalability of ABR chemistries and optimize manufacturability.
 - Enable the development of a domestic supply chain for large format battery manufacturing.

Partners








- Interactions/Collaborations
 - National Laboratories: ANL, SNL, NREL
 - Battery Manufacturers: XALT Energy, A123 Systems, Navitas Systems
 - Material/Process Suppliers: PPG Industries, Phostech Lithium, TODA America, Superior Graphite, Zenyatta Ventures, Timcal, JSR Micro, Solvay Specialty Polymers, Vorbeck, XG Sciences, FMC Lithium
 - Equipment Manufacturer: Frontier Industrial Technology, Miltec UV, Conquip, BTU International, Sharp Laboratories of America
- Project Lead: ORNL

Relevance & Objectives

- Main Objective: Demonstrate scalability and manufacturability of ABR technology and enable a domestic supply chain for large format battery manufacturing.
 - Include ABR active materials in large scale manufacturing studies
 - Process quantities of electrodes for research use with collaborators and ABR PIs
 - Optimize manufacturing processes when scaling
 - Demonstrate large scale applicability and benchmark materials
 - Advise equipment manufacturers and enable domestic supply chain
- Relevance to Barriers and Targets
 - Scalability of new materials and manufacturing procedures are not trivial
 - Cost calculations are currently including insufficient details about manufacturing cost and energy consumption
 - Manufacturing procedures, formation cycle protocols, and drying operations are currently highly proprietary and led by Asian suppliers
 - Implementation of low-cost, green manufacturing methodology for lithium ion battery electrodes across all manufacturing steps (to meet \$300/kWh 2015 VTO storage goal for PHEV-40s).
 - **Preserve** long-term performance: achieve a lifetime of 10 years and 1000 cycles at 80% DOD for EVs and 5000 deep discharge cycles for PHEVs.



Project Milestones

Status	Milestone or Go/No-Go	Description
Complete 	FY13 Q1	Successful installation and startup of facility and successful operation well above specified performance (dew point spec: $<-30^{\circ}\text{C}$, achieved: $<-56^{\circ}\text{C}$).
Complete 	FY13 Q2	Successful processing and performance comparison of tape-cast and slot-die electrode coatings.
Complete 	FY13 Q3	Determined maximum electrode stack number and capacity in 12 mm thick pouch cell with ABR cathode reference NCM 532 and anode reference graphite A10/12 and demonstration of assembly issues with pouches beyond 10 mm thickness.
Complete 	FY13 Q4	Provide 100ft of electrodes or more to ABR collaborators.
Complete 	FY14 Q1	Obtain cell-level confidence intervals (performance variability at 0.2C, 1C, and 5 C discharge rates) on 1-Ah pouch cell production, and determine monthly cell production rate of the ORNL BMF.
Complete 	FY14 Q2	Produce minimum of 500 ft of ABR baseline anode (CPA12 graphite).
Complete 	FY14 Q3	Produce minimum of 500 ft of ABR baseline cathode (TODA NMC 532).
9/2014	FY14 Q4 – Go/No-Go	Provide at least 500 ft of ABR baseline anode and cathode for use in various ABR projects according to VTO guidance.

Project Approach

Utilization of DOE Battery Manufacturing R&D Facility (BMF) at ORNL

funded by DOE Vehicle Technologies Program and Advanced Manufacturing Office

- Fully integrated roll to roll operation.
- Automated single steps to guarantee maximum reliability and repeatability.
- Manual material handling from station to station to allow for maximum flexibility.
- Pouch cells of up to 55 x 85 x 12 mm and 6-7 Ah
- Focus on manufacturing R&D, drying, alternative heating technologies, solventless processing, alternative assembly methods, new cutting, materials handling, and filling methods.
- Production yield issues
- 700 sqft dry space with <0.5% R.H.
- 700 sqft adjustable 1-15% R.H.
- Located in manufacturing R&D park together with additive manufacturing, room-temperature materials synthesis, roll-to-roll processing, direct powder feed rolling, robotics R&D
- In easy to access off-site location specifically designed for industrial collaborations.



Project Approach

Building a U.S. supply chain and competitive industry

- ORNL provides a one of a kind leadership facility for industry to collaborate
- Access and IP protected collaboration
- Resource for
 - Chemical and materials suppliers
 - Battery manufacturers and their customers
 - System integrators
 - Original equipment manufacturers
- Development of processes, process optimization, manufacturing schemes, materials improvements, diagnostics, and production yield



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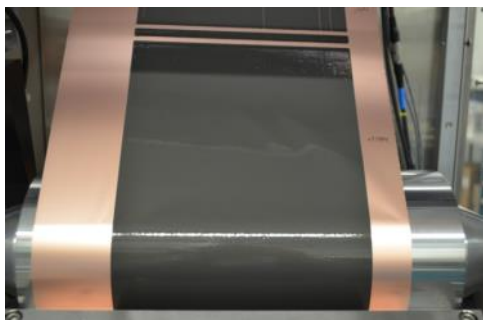


Technical Accomplishments – Electrode scaling and manufacturability

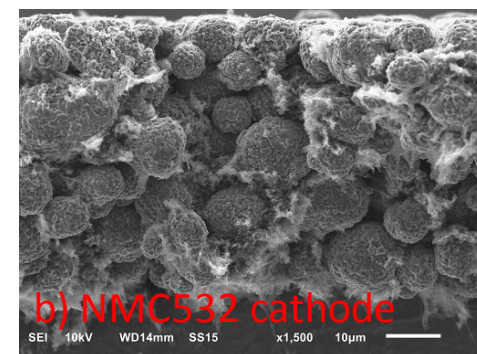
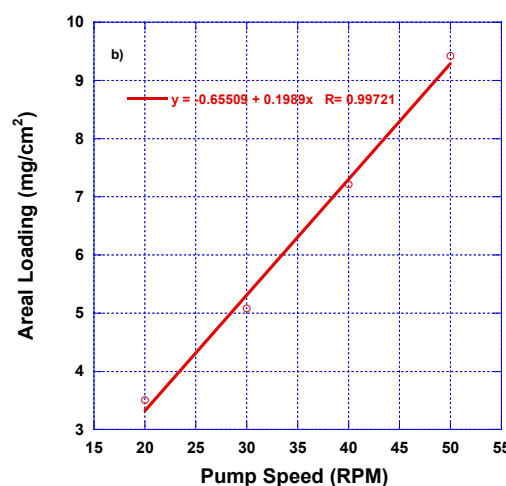
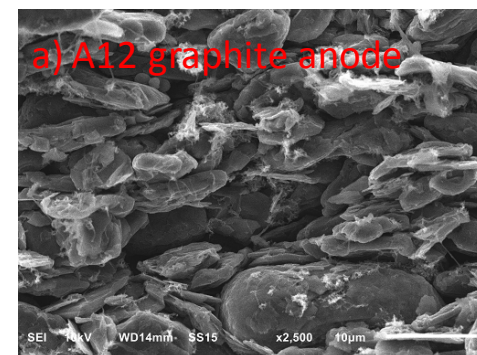
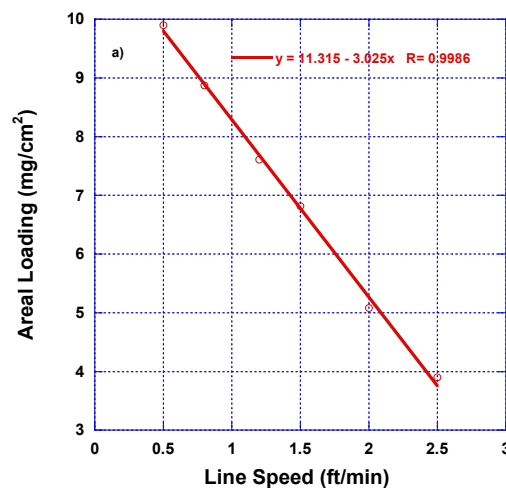
- FY13
 - A12 graphite baseline anode: ~400 ft, 6" wide; 10 ft each sent to ANL and SNL for round robin testing; 10 ft sent to NREL for IR image and porosity test; 100 ft for CERC-CVC project; the rest used by BMF activities
 - NMC532 baseline cathode: ~400 ft, 6" wide; 10 ft each sent to ANL and SNL for round robin testing; 10 ft sent to NREL for IR image and porosity test; 100 ft for CERC-CVC project; the rest used by BMF activities
 - HE5050 coating: ~80 ft, 6" wide, for neutron diffraction experiment
 - LFP coating through aqueous processing: ~60 ft, 6" wide for ABR projects
 - NMC532 coating through aqueous processing: ~150 ft, 8" wide for 3 different water soluble binders; for ABR projects
 - LCO coating for Plextronics: ~50 ft, 8" wide; for WFO
 - NMC333 coating for Plextronics: ~250 ft, 8" wide for 5 different Plexcore contents; for WFO
 - NCA coating for Plextronics: ~250 ft, 8" wide for 5 different Plexcore contents for WFO
- FY14:
 - NMC532 with latex binder via aqueous processing: ~40 ft, 8" wide; ~1 ft sent to Enerize; the rest for ABR projects
 - NMC532 with SBR binder via aqueous processing: ~40 ft, 8" wide for ABR projects
 - NMC532 with TRD202A binder via aqueous processing: ~140 ft, 8" wide; ~40 ft sent to ANL for thickness study; the rest for ABR projects
 - NMC532 with Nafion® binder via aqueous processing: ~40 ft, 8" wide for ABR projects
 - A12 graphite with latex binder via aqueous processing: ~40 ft, 8" wide; ~1 ft sent to Enerize
 - A12 graphite with SBR binder via aqueous processing: ~40 ft, 8" wide; ~5 ft sent to NREL for porosity measurement; the rest for ABR projects
 - A12 graphite with TRD102A binder via aqueous processing: ~140 ft, 8" wide; ~40 ft sent to ANL for thickness study; ~5 ft sent to NREL for porosity measurement; ~1 ft sent to Enerize
 - NMC532 baseline coating: ~200 ft, 8" wide; ~30 ft for ORNL TIP project; ~10 ft sent to NREL for porosity measurement; ~2 ft sent to Enerize; the rest for ABR projects
 - A12 graphite baseline coating: ~200 ft, 8" wide; ~60 ft for MDF project with Vorbeck; ~1 ft sent to Enerize; the rest for ABR projects
 - Superior graphite 1520 P baseline coating: ~40 ft, 8" wide for ABR projects
 - Superior graphite 1512 P baseline coating: : ~40 ft, 8" wide for ABR projects
 - Graphite coating: ~30 ft, 8" wide for ORNL TIP project
 - NMC coating for Vorbeck: ~60 ft, 8" wide for Vorbeck project

2,570ft of high quality electrodes

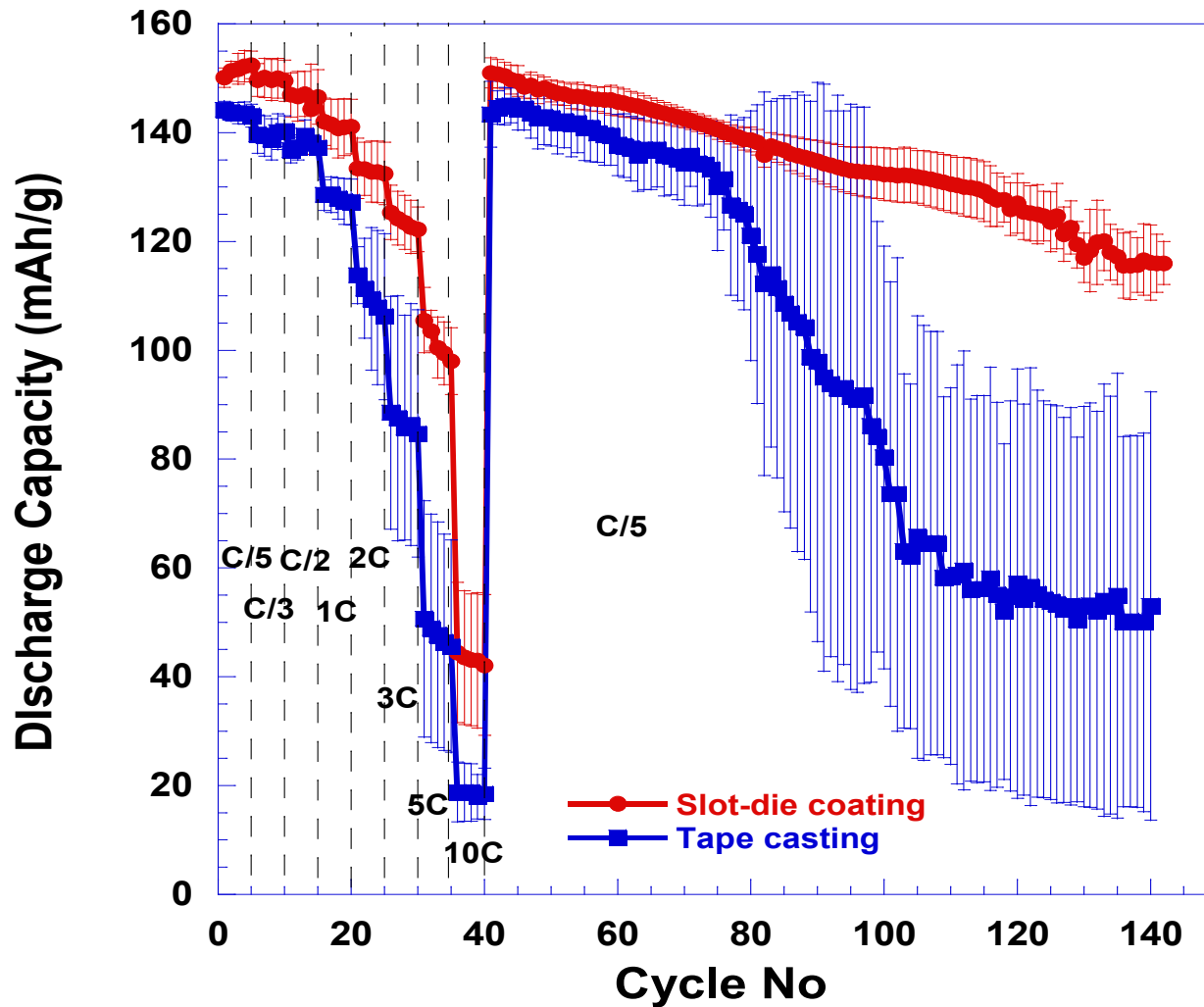
Coating quality and matching materials loading



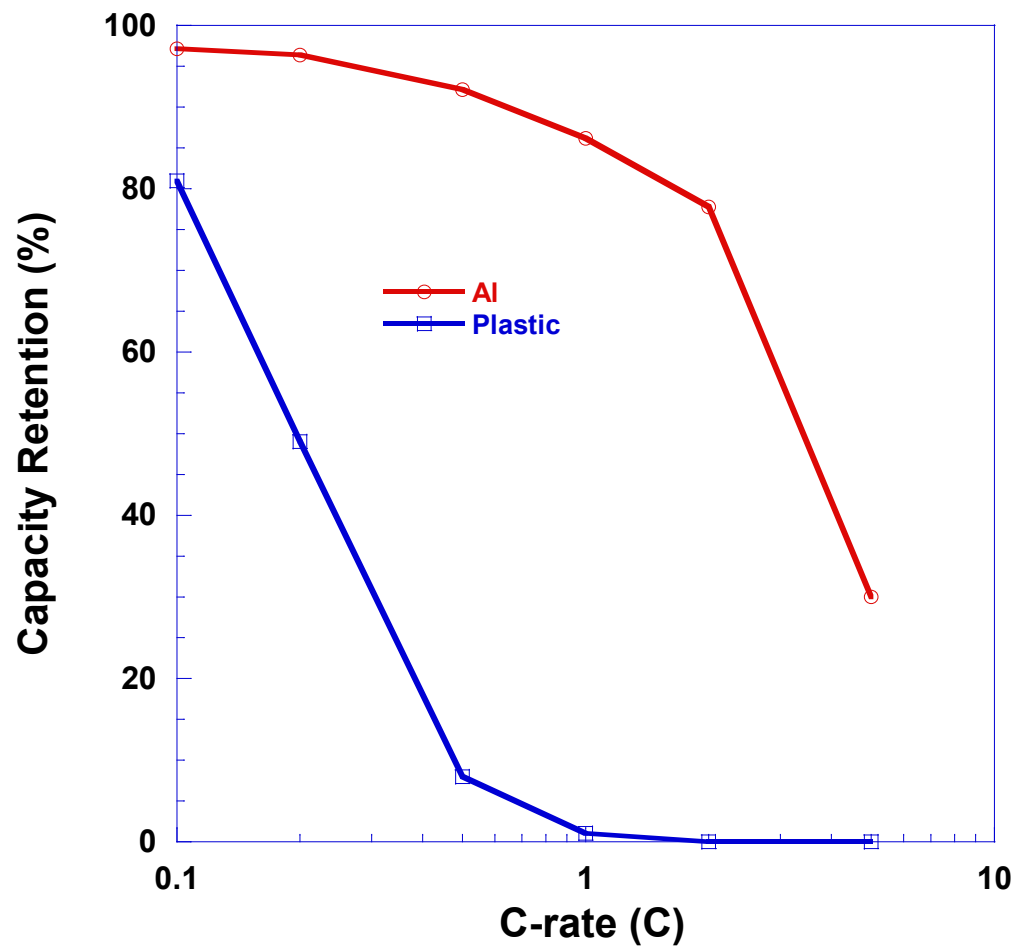
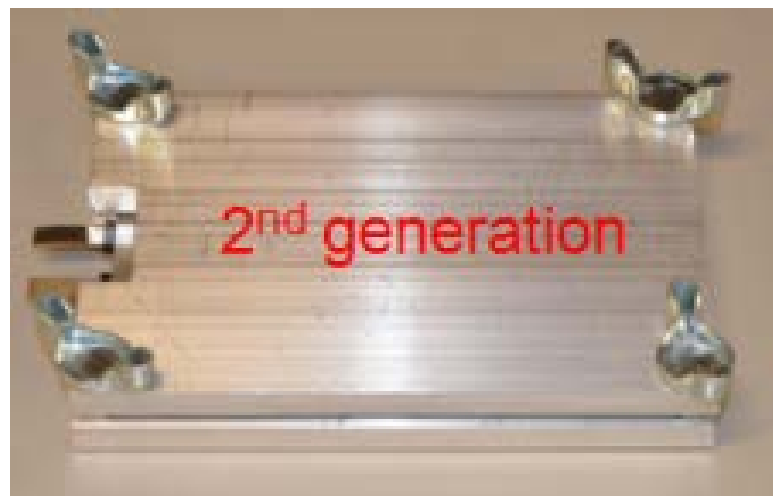
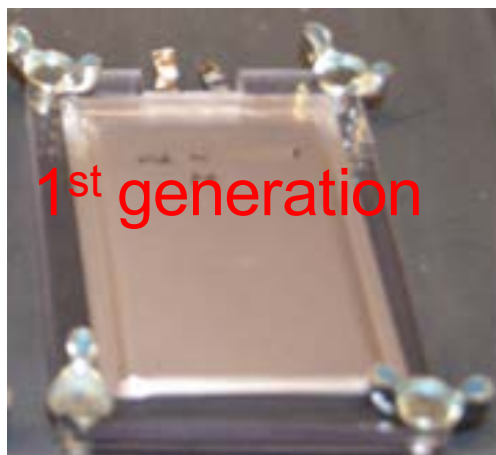
Electrode	Composition	Areal loading (mg/cm ²)	Porosity (%)
A12	A12/Super P Li/9300 PVDF =92/2/6 wt	7.51	52.2
NMC532	NMC532/Denka carbon black/ 5130 PVDF =90/5/5 wt	12.57	49.0



Slot-die coating outperforms tape cast coating



Test standards

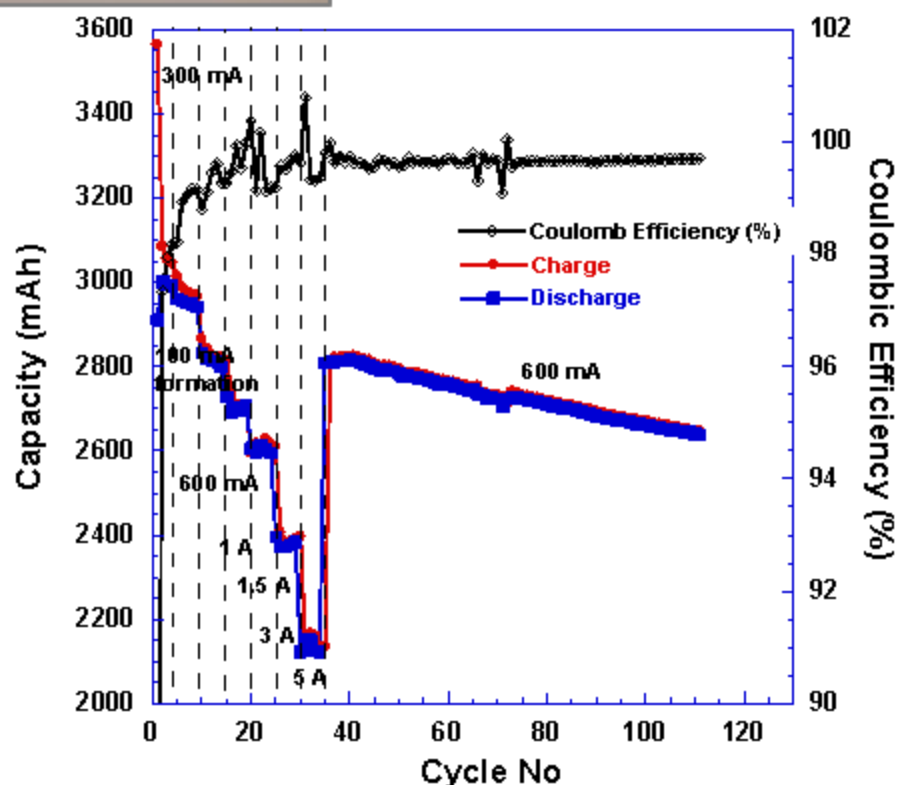
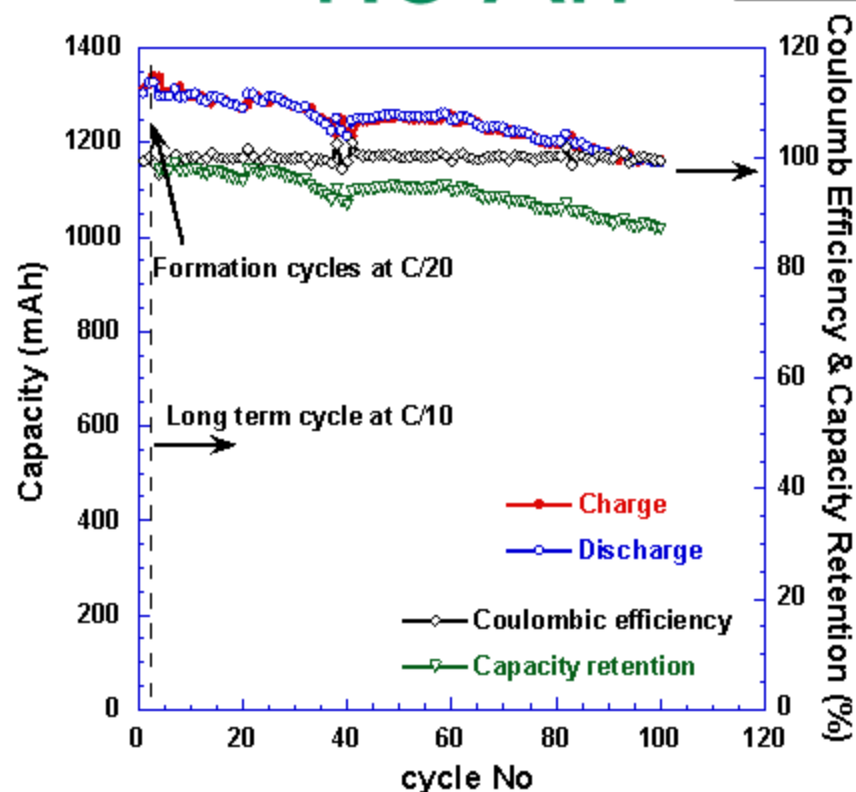


Large cell demonstration

1.3 Ah

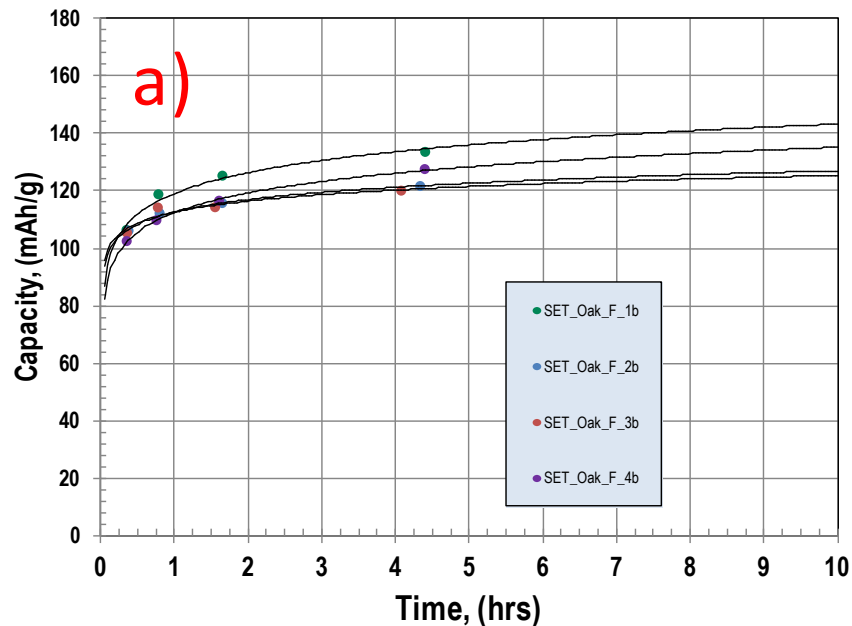


3 Ah

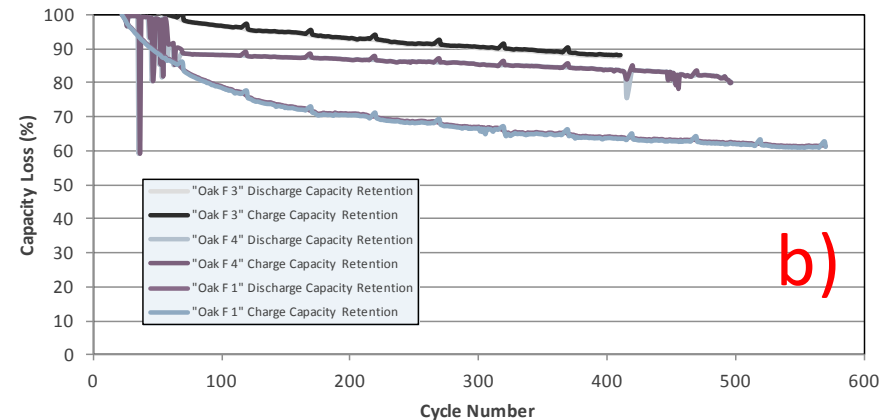


Performance testing of ORNL electrodes at ANL

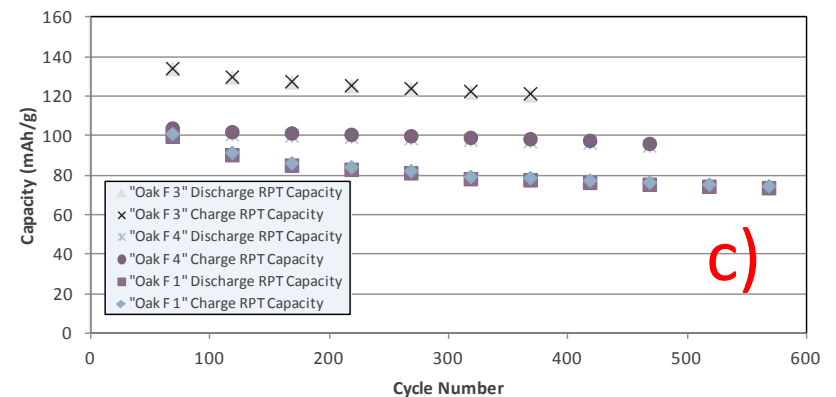
Full Cell Rate Study



Capacity Loss vs Cycle

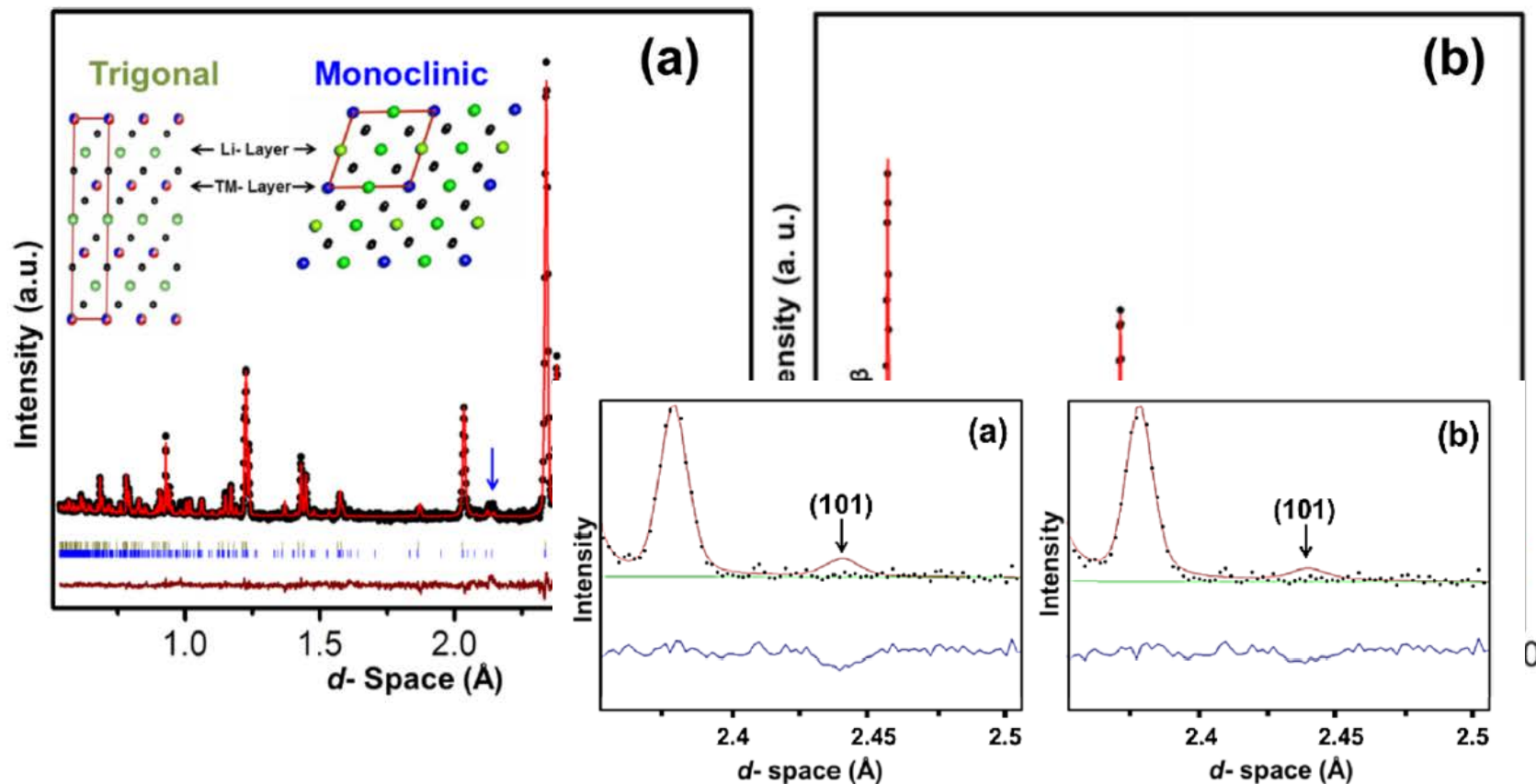
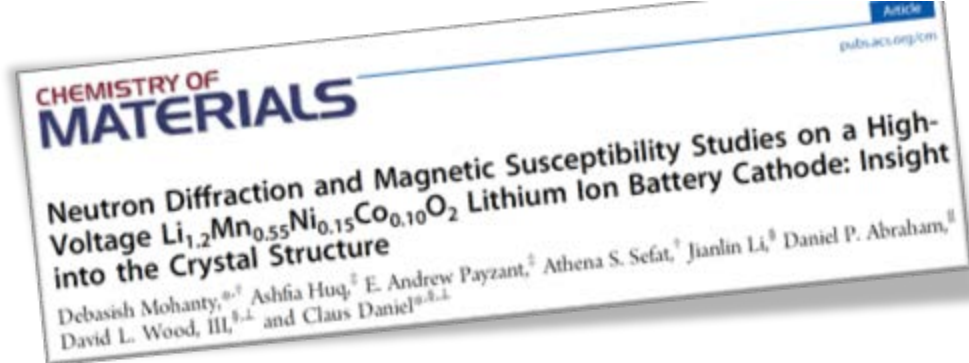


RPT Capacity (mAh/g) vs Cycle Number



ORNL contributions to LMR-NMC studies

More info: see LMR-NMC session



Cycled materials for neutron diffraction experiment at POWGEN @ ORNL

- one gram of material is needed to obtain better counts



Pouch cell with 3 unit cells

Pouch cells of dimensions: 84.4 mm long & 56.0 mm wide

Electrode solid loading ~ 10 mg/cm²

Cathode (positive electrode): Toda HE5050 LMR-NMC

Anode (negative electrode): Round robin baseline graphite A12

Homogeneous electrodes were fabricated by Slot-die coater

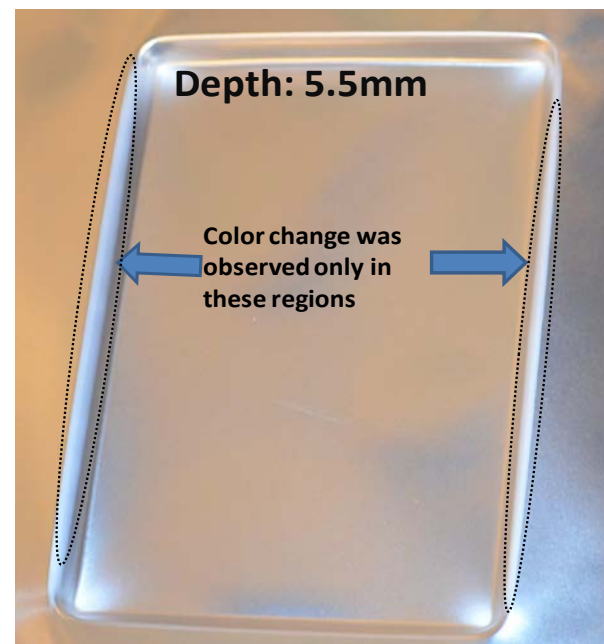


Manufacturing limitations

Gas formation



Deep draw limitation



Collaborations

- Partners

- National Labs: Argonne National Laboratory, Sandia National Laboratories
- Battery Manufacturers: Dow Kokam, A123 Systems, Navitas Systems
- Active Material Suppliers: Phostech Lithium, TODA America, Superior Graphite, Zenyatta Ventures
- Inactive Material Suppliers: JSR Micro, Solvay Specialty Polymers, Timcal, XG Sciences
- Equipment/Coating Suppliers: PPG Industries, Frontier Industrial Technology



- Collaborative Activities

- Provide access to manufacturing facilities, expertise, and scale up
- Enable domestic supply chain by being unbiased advisor and independent facilitator
- Benchmark against commercially available materials
- Scale-up logistics and manufacturing cost savings of processing with key battery developers and raw materials suppliers.

Leveraging activities

ABR program, CERC

AMO

WFO and user projects



EERE Roll-to-roll and NDE workshop

Future Work

- Remainder of FY14
 - Accomplish all milestones and provide materials to collaborators
 - Continue to assemble pouch cells and evaluate new materials
- FY15
 - Widen collaborator network
 - Standardize processing and manufacturing procedures
 - Include new chemistries from ABR and BATT
 - Provide scaling expertise for renewable carbon materials, silicon materials, sulfur, thick electrodes, reducing solvents

Information Dissemination and Commercialization

- Selected contributions out of over 40 in FY13/14

- Selected publications

1. J. Li, B.L. Armstrong, J. Kiggans, C. Daniel, and D.L. Wood, "Lithium Ion Cell Performance Enhancement Using Aqueous LiFePO_4 Cathode Dispersions and Polyethyleneimine Dispersant," *Journal of The Electrochemical Society*, **160**, A201–A206 (2013).
2. J. Li, B.L. Armstrong, J. Kiggans, C. Daniel, and D.L. Wood, "Optimization of Multicomponent Aqueous Suspensions of LiFePO_4 Nanoparticles and Carbon Black for Lithium Ion Battery Cathodes," *Journal of Colloid and Interface Science*, **405**, 118–124 (2013).
3. D.L. Wood, C. Daniel, and J. Li, "Prospects for Reducing the Cost of Lithium Ion Electrode Processing and Pouch Cell Formation Steps," *Journal of Power Sources*, In Preparation, 2014.
4. D. Mohanty, S. Kalnaus, R.A. Meisner, K.J. Rhodes, E.A. Payzant, D.L. Wood, and C. Daniel, "Structural Transformation of a Lithium-Rich $\text{Li}_{1.2}\text{Co}_{0.1}\text{Mn}_{0.55}\text{Ni}_{0.15}\text{O}_2$ Cathode During High Voltage Cycling Resolved by In-Situ X-Ray Diffraction," *Journal of Power Sources*, **229**, 239–248 (2013).
5. D. Mohanty, S. Kalnaus, R.A. Meisner, A. Safa-Sefat, J. Li, K.J. Rhodes, E.A. Payzant, D.L. Wood, and C. Daniel "Structural Transformation in a $\text{Li}_{1.2}\text{Co}_{0.1}\text{Mn}_{0.55}\text{Ni}_{0.15}\text{O}_2$ Lithium-Ion Battery Cathode During High-Voltage Hold," *RSC Advances*, **3**, 7479–7485 (2013).
6. D. Mohanty, A. Safa-Sefat, S. Kalnaus, J. Li, R.A. Meisner, E.A. Payzant, D.P. Abraham, D.L. Wood, and C. Daniel, "Investigating Phase Transformation in $\text{Li}_{1.2}\text{Co}_{0.1}\text{Mn}_{0.55}\text{Ni}_{0.15}\text{O}_2$ Lithium-Ion Battery Cathode During High-Voltage Hold (4.5 V) via Magnetic, X-ray Diffraction and Electron Microscopy Studies," *Journal of Materials Chemistry A*, **1**, 6249–6261 (2013).
7. D. Mohanty, A. Huq, E.A. Payzant, A. Safa-Sefat, J. Li, D.P. Abraham, D.L. Wood, and C. Daniel, "Neutron Diffraction and Magnetic Susceptibility Studies on a High-Voltage $\text{Li}_{1.2}\text{Mn}_{0.55}\text{Ni}_{0.15}\text{Co}_{0.1}\text{O}_2$ Lithium-Ion Battery Cathode; an Insight to the Crystal Structure," *Chemistry of Materials*, **25**, 4064–4070 (2013).
8. D. Mohanty, A. Safa-Sefat, J. Li, R.A. Meisner, A.J. Rondinone, E.A. Payzant, D.P. Abraham, D.L. Wood, and C. Daniel, "Correlating Cation Ordering and Voltage Fade in a Lithium-Manganese-Rich Layered-Layered Lithium-Ion Battery Cathode Oxide; a Joint Magnetic Susceptibility and TEM Study," *Physical Chemistry Chemical Physics*, **15**, 19496–19509 (2013).
9. D. Mohanty, J. Li, R. Born, L.C. Maxey, R.B. Dinwiddie, C. Daniel, and D.L. Wood, "Non-Destructive Evaluation of Slot-Die-Coated Lithium Secondary Battery Electrodes by In-Line Laser Caliper and IR Thermography Methods," *Analytical Methods*, **6**, 674–683 (2014).
10. D. Mohanty, J. Li, A. Safa-Sefat, A. Huq, D.P. Abraham, E.A. Payzant, D.L. Wood, and C. Daniel, "Layer to Spinel Transformation Mechanisms in a Lithium-Rich High-Voltage Lithium-Ion Battery Cathode," *Advanced Energy Materials*, Submitted, 2014.

- Selected presentations

1. D. Wood, J. Li, D. Mohanty, S. Kalnaus, B. Armstrong, C. Daniel, and B. Brown, "Advanced Materials Processing and Novel Characterization Methods for Low-Cost, High Energy-Density Lithium-Ion Batteries," Advanced Automotive Battery Conference 2013, Pasadena, California, February 4-8, 2013. (Invited).
2. C. Daniel, D. Wood, J. Li, B. Armstrong, J. Kiggans, D. Mohanty, and S. Kalnaus, "Electrification of Transportation – Cost and Opportunities", Bridging the Gap Conference 2013, Oak Ridge, Tennessee, March 5-6, 2013 (Invited).
3. J. Li, D. Mohanty, B. Brown, C. Daniel, and D. Wood, "Fabrication and Performance of $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ Cathodes through Aqueous Processing with Various Binders," Advanced Automotive Battery Conference 2014, Atlanta, Georgia, February 4, 2014. (Invited).
4. J. Li, D. Mohanty, C. Daniel, and D. Wood, "Aqueous Processing of $\text{LiNi}_{0.5}\text{Mn}_{0.3}\text{Co}_{0.2}\text{O}_2$ Composite Cathodes for Lithium-Ion Batteries," 225th Meeting of The Electrochemical Society, Orlando, Florida, Abstract No. 194, May 11-16, 2014.
5. D. Mohanty, J. Li, C.L. Maxey, R.B. Dinwiddie, C. Daniel, and D. Wood, "In-Line Non-Destructive Testing of a Lithium-Ion Battery Electrode by Laser Caliper and Thermography," 2013 MRS Fall Meeting & Exhibit, Boston, Massachusetts, December 1-6, 2013.
6. D. Mohanty, J. Li, A. Huq, E.A. Payzant, D.L. Wood, III, and C. Daniel, "Understanding Voltage Fade Mechanism in a Lithium and Manganese Rich Layered-Layered High-Voltage Lithium-Ion Battery Cathode by Neutron Diffraction Studies," 2013 MRS Fall Meeting & Exhibit, Boston, Massachusetts, December 1-6, 2013.
7. D. Mohanty, J. Li, D.L. Wood, III, and C. Daniel, "Understanding the Structural Transformation in High-Voltage $\text{Li}_{1.2}\text{Mn}_{0.55}\text{Ni}_{0.15}\text{Co}_{0.1}\text{O}_2$ Lithium-Ion Battery Cathode via Neutron Diffraction and Magnetic Susceptibility Studies," 2014 MRS Spring Meeting & Exhibit, San Francisco, California, April 21-25, 2014.
8. C. Daniel, Current and Future Battery Chemistries and Large-Format Cells, National Academy of Engineering, U.S. Frontiers of Engineering Symposium, Irvine, CA, September 2014

Acknowledgements

- U.S. DOE Office of Energy Efficiency and Renewable Energy (EERE) Vehicle Technologies Office (Program Managers: David Howell and Peter Faguy)
- ORNL Contributors:
 - Claus Daniel
 - David Wood III
 - Jianlin Li
 - Beth Armstrong
 - Brad Brown
 - Debasish Mohanty
 - Shrikant Nagpure
- Technical Collaborators:
 - Andrew Jansen
 - Bryant Polzin
 - Chris Orendorff
 - David Telep
 - Erin O'Driscoll
 - Maneesh Bahadur
 - Mike Wixom
 - James Banas
 - Gregg Lytle
 - Jack Burgman
 - Stuart Helling



Summary

- **Objective:** Demonstrate scalability and manufacturability of ABR technology and enable a domestic supply chain for large format battery manufacturing
- **Approach:** Utilize DOE Battery Manufacturing R&D Facility (BMF) at ORNL for extensive large scale manufacturability study
 - Extensive equipment
 - Industrial collaborations with extensive WFO portfolio to understand industry needs
 - Extensive expertise and capabilities
- **Technical:**
 - Demonstrate coating and processing in continuous 8" wide roll to roll operation on 12" web
 - Scale pouch cells from 150mAh to 3Ah
 - Control process and repeatability
 - Test processing limits
- **Collaborators:** Extensive collaborations with national laboratories, lithium-ion battery manufacturers, raw materials suppliers, and coating producer.
- **Commercialization:** Highly engaged with industry and all ABR PIs; high likelihood of technology transfer because equipment compatibility and understanding of needs.