

Dry Process Electrode Fabrication

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Project ID: ES134

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Timeline

- Project start date: Oct. 2011
- Project end date: Oct. 2015
- Percent complete: xx%

Budget

- **Total project funding: \$4,239,879**
 - Government Share: \$2,992,744
(DOE Obligations thru 4/3/2014 \$1,643,565)
 - Contractor Share: \$1,247,135
- **Expenditures of Gov't Funds:**
 - FY13: \$534,530 (10/1/2012-09/30/2013)
 - FY14: \$371,391 (10/1/2013- 03/31/2014)
 - Total: \$1,137,946 (thru 3/31/2014)

Barriers

- Conventional slurry casting processes drive the cost of lithium ion battery electrodes.

Partners

- No other funded partners
- Non-funded collaborators listed on slide xx.

Objectives and relevance of this project



- The Phase I objectives of this project are:
 - + PTFE binders have been demonstrated for solvent-free cathodes, but PTFE is not electrochemically stable in a lithium battery anode. Therefore phase I will define a binder system for solvent-free anode fabrication that is stable over 500 cycles to full state of charge.
 - + Identify the thickness limit for dry process cathodes that can meet EV rate and cycle life criteria
- The Phase II objectives of this project are:
 - + Produce a solvent-free anode material that capacity matches the Phase I cathode.
 - + Produce free standing dry process cathode that retains 50% capacity at 1C rate.
 - + Validate cost model by running pilot coating line.
 - + Deliver 24 cells in SOA EV cell format.

Project Milestones and Decision Points



Milestone/Decision Point	Metric	Date
1. Acceptance of mgt plan revisions		√
2. Down-select LMFP, NMC, and pre-coat		√
3. Cathode morphology and mixing conditions specified		√
4. High solid loading anode	>40% solids cast to >3 mAh/cm ²	√
5. Demo. lab prototype cell w/ dry process blended cathode	>100 μm cathode	√
6. Deliver interim cells with dry process blended cathode/wet anode	14 cells, 4 Ah pouch	√
7. Demo dry process anode	Rate/capacity match cathode	Oct. 14
8. Down-select low cost anode process	50% vs baseline capex + opex	Dec. 14
9. Scale cathode film to support task 16	10 m	Apr. 15
10. Lab prototype cell dry anode/dry cathode	Pass EV life test	June 15
11. Deliver final cells	24 cells, >14 Ah prismatic can	Sep. 15

Approach



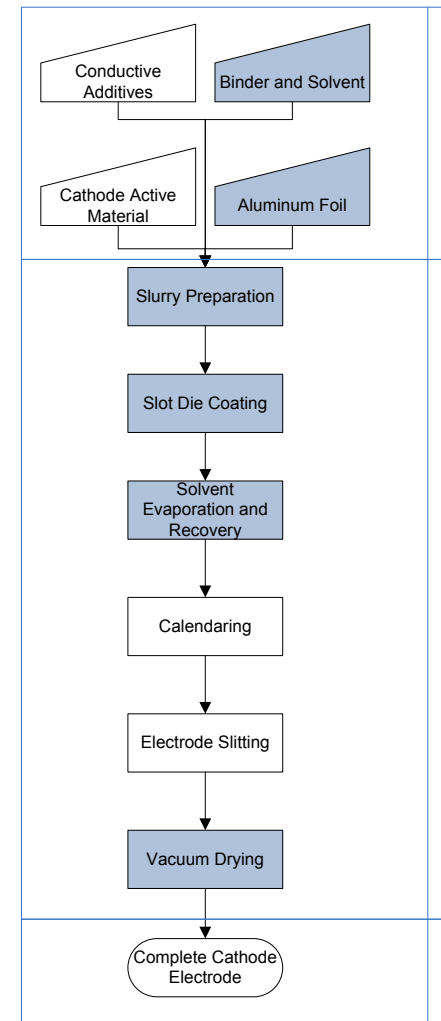
First, optimize blend of phosphate and oxide cathode powders with respect to energy, safety and compatibility with dry electrode fabrication.

Second, modify low- or zero-solvent binders for anodic compatibility.

Third, down-select cathode and anode formulations. Scale up processes to support prototype cell assembly and cost model validation.

Validate performance and life in EV-relevant prototype cells.

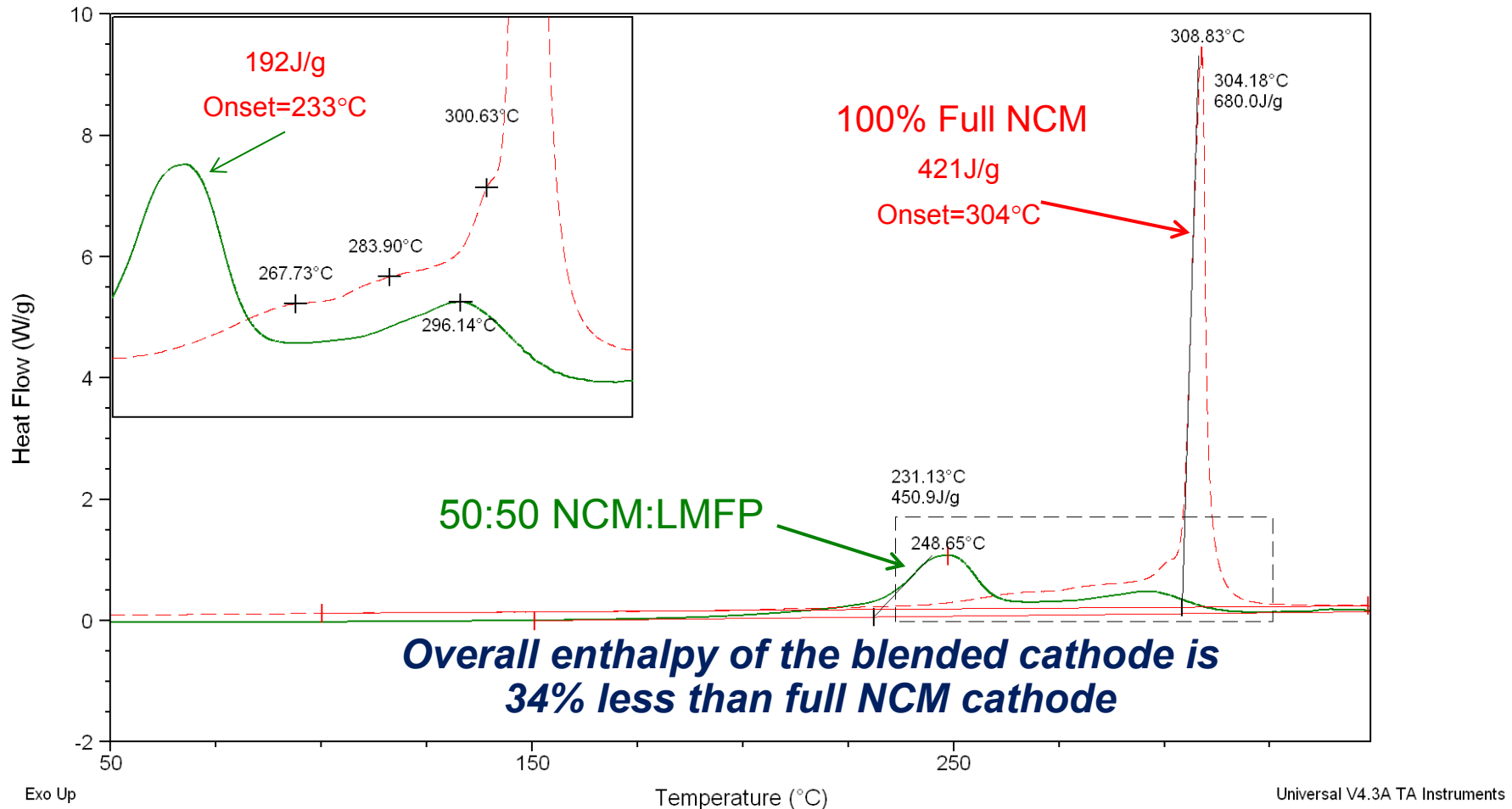
The dry fabrication process reduces cost by eliminating the shaded steps required in conventional slurry cast electrode fabrication.



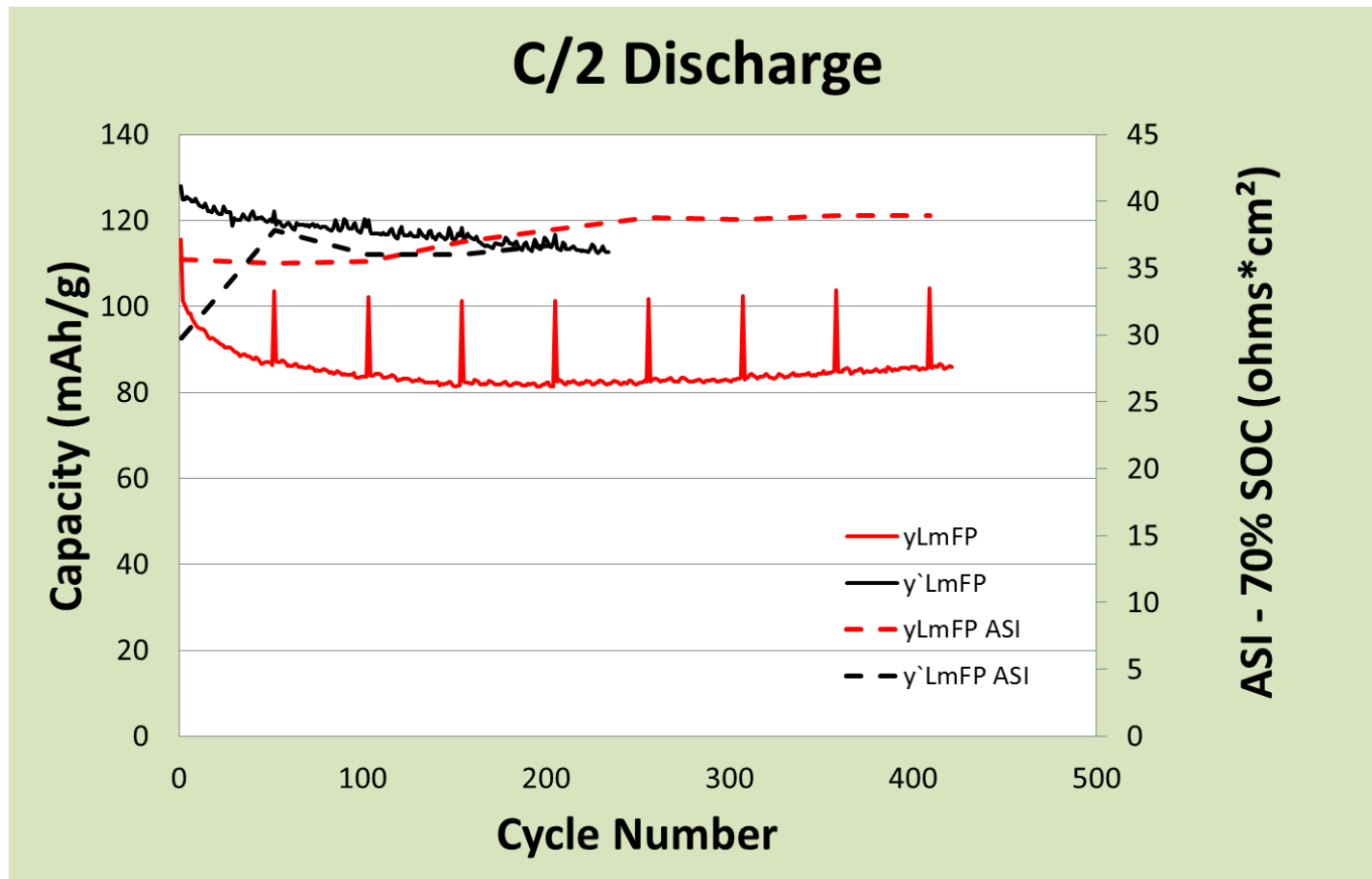
Cathode blending improved safety and processability.



The blended dry processed cathode appears to suppress the high exothermic peak observed at ~309°C for the full NCM cathode



Full cell cycle life data for dry process cathodes.



New mixing process (black) improved electrode capacity and DCR vs. Q3 reported results (red). Attributed to improved dispersion of conductive additive. The y' (black) electrode formulation details are provided in table on slide 13. Cycle life data from double layer pouch full cell.

Anode option 1 : Advance Drying Process (ADP) for high throughput coating.



Status:

- Developed high solids aqueous anode slurry
- Adapted an emerging drying/curing technology, derived from commercial curing and annealing operations in the semiconductor industry
- Worked with the leading process equipment developer in this area to validate the process for low-cost anode production
- Demonstrated that the ADP can reduce coating drying time by more than 50% without compromising the anode quality

Indication:

- A rapid drying coating process is achievable in a conventional slot-die coater by applying the newly developed high solids aqueous slurry
- The ADP method dries aqueous anode coating rapidly at even lower temperature
- It might be a very promising solution to developing a low energy consuming and high throughput anode coating process by integrating ADP to a slot-die coater

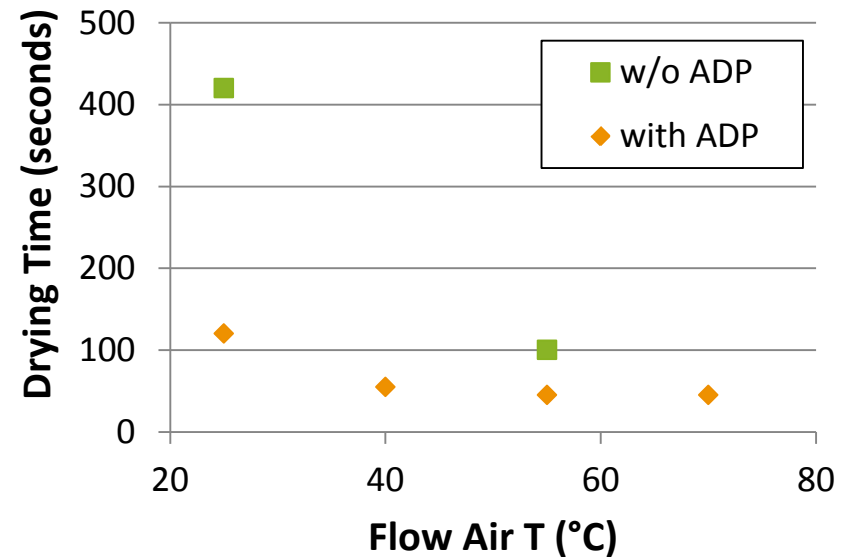
Advance Drying Process (ADP) for high throughput low cost anode coating



Anode coating drying time with or without ADP

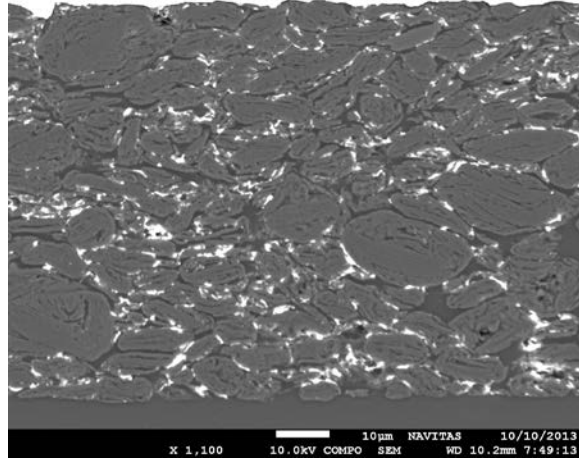
Drying T (°C)	ADP (on/off)	Drying Time (Second)
25	off	420
25	on	120
40	on	55
55	off	100
55	on	45

Anode coating drying time with or w/o ADP

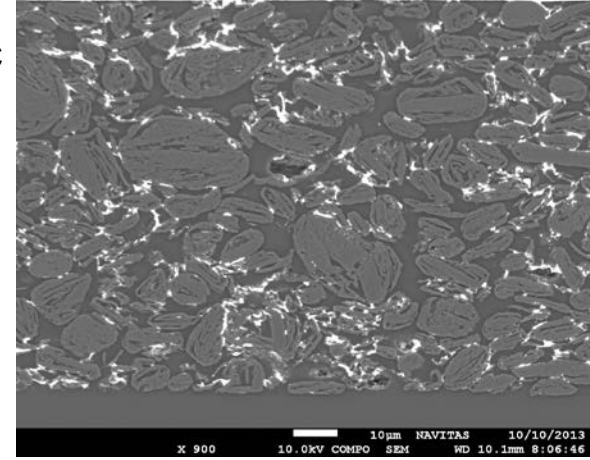


- With ADP, the drying time was reduced from 420 seconds to 120 seconds at 25°C and from 100 seconds to 45 seconds at 55°C

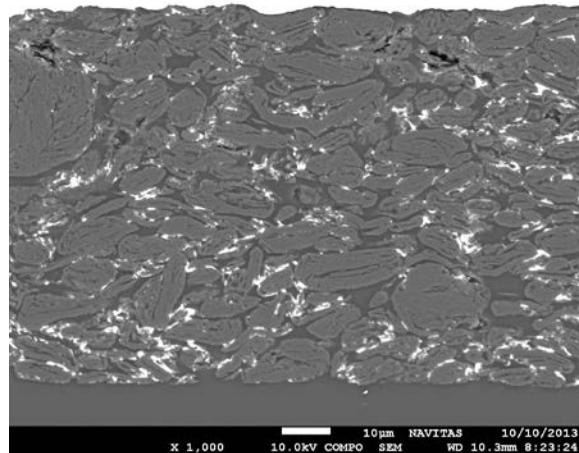
Advance Drying Process (ADP) for high throughput low cost anode coating



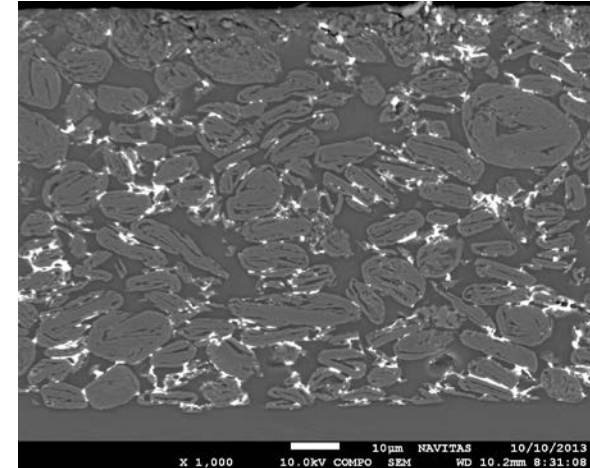
25°C
air
with
ADP



25°C
air



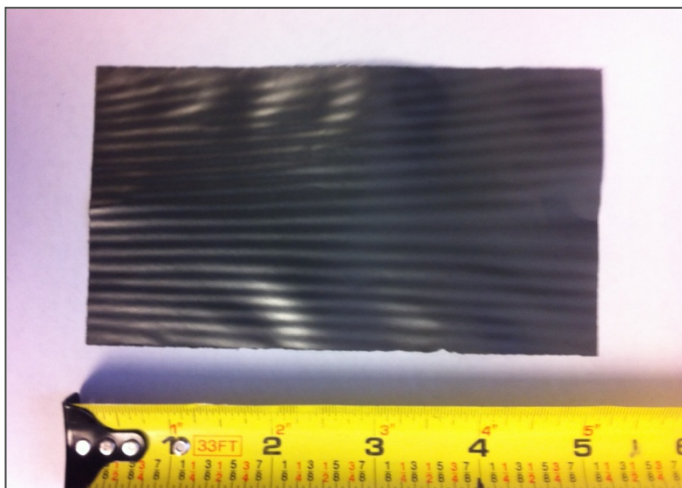
55°C
air
with
ADP



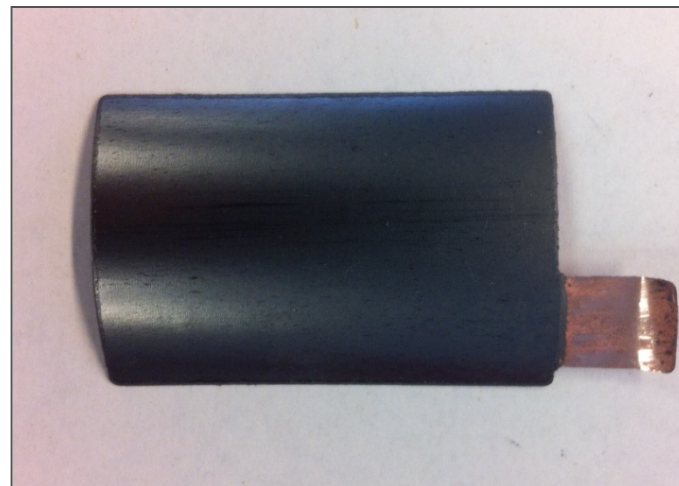
55°C
air

- Although the drying time is much shorter, the ADP dried samples (left) show no difference from the baseline (right) binder distribution. (Binder stained to appear as white).

Anode option 2: Solvent-free anode fabrication



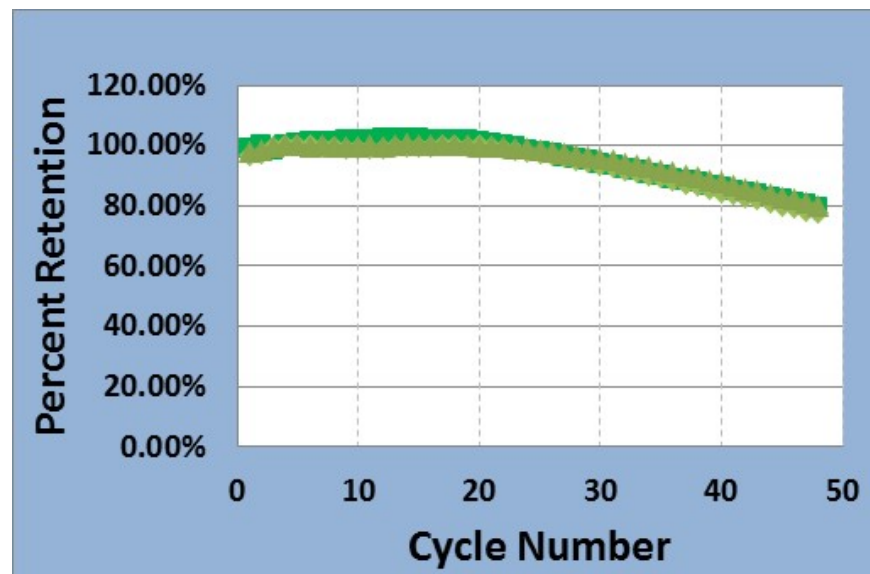
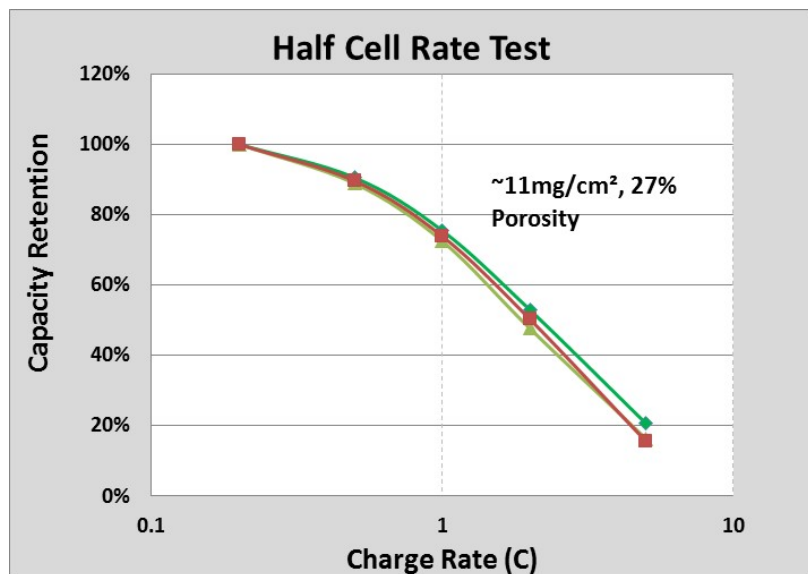
**Flexible free standing ~ 90 μm
anode film (dry process)**



**Anode with good adhesion
(laminating the film to Cu foil)**

- Dry blend graphite, carbon, and polymer binder powder to form a homogeneous powder mixture
- Calender the mixture to form a flexible film
- Laminate the film to current collector (with calendering) to form an anode
- Sample evaluation will be performed

Solvent-free anode performance



Capacity degradation after 25 cycles is attributed to binder failure. 2014 work is developing binder blends that reduce ICL and improve cycle life.

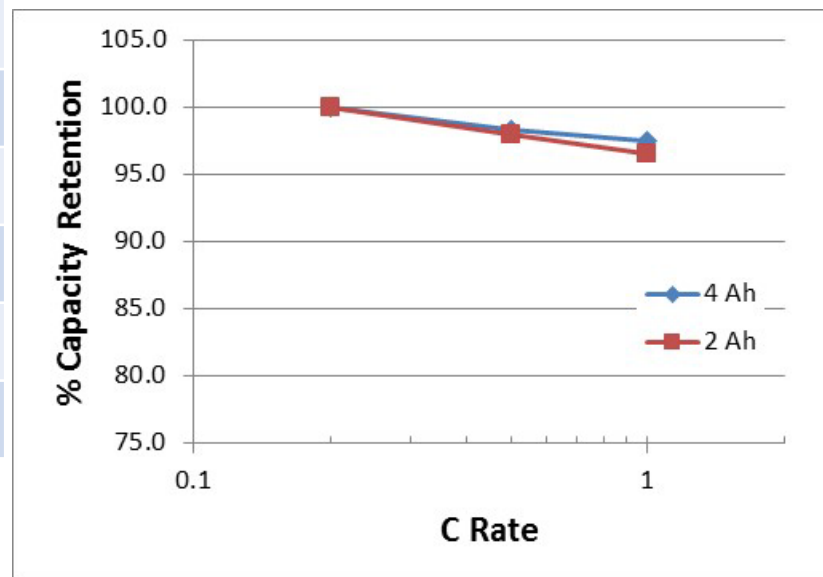
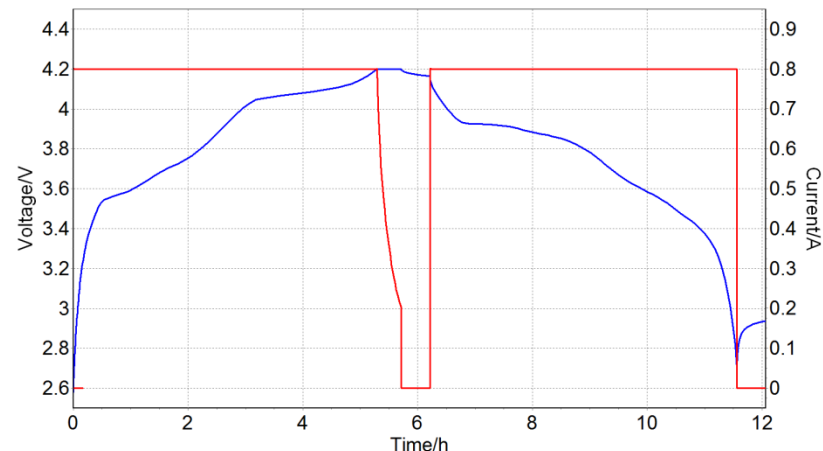
Interim prototype cell deliverable specifications



Interim Deliverable Summary

Cathode formulation	82% active (50:50 NMC:LMFP) 5% activated C 5% conductive C 8% PTFE binder	
Cathode loading	23 mg/cm ²	
Cathode porosity	36 %	
Cathode thickness	230 μm*	
Cell capacity (Ah at C/5)	4.3	2.1
% capacity at 1C	97	97
Cell dimensions (mm)	57 x 105 x 8	57 x 105 x 4.5
Cell mass (g)	92	50
Reversibility	79	78

* Double-sided, incl 20 μm Al foil current collector

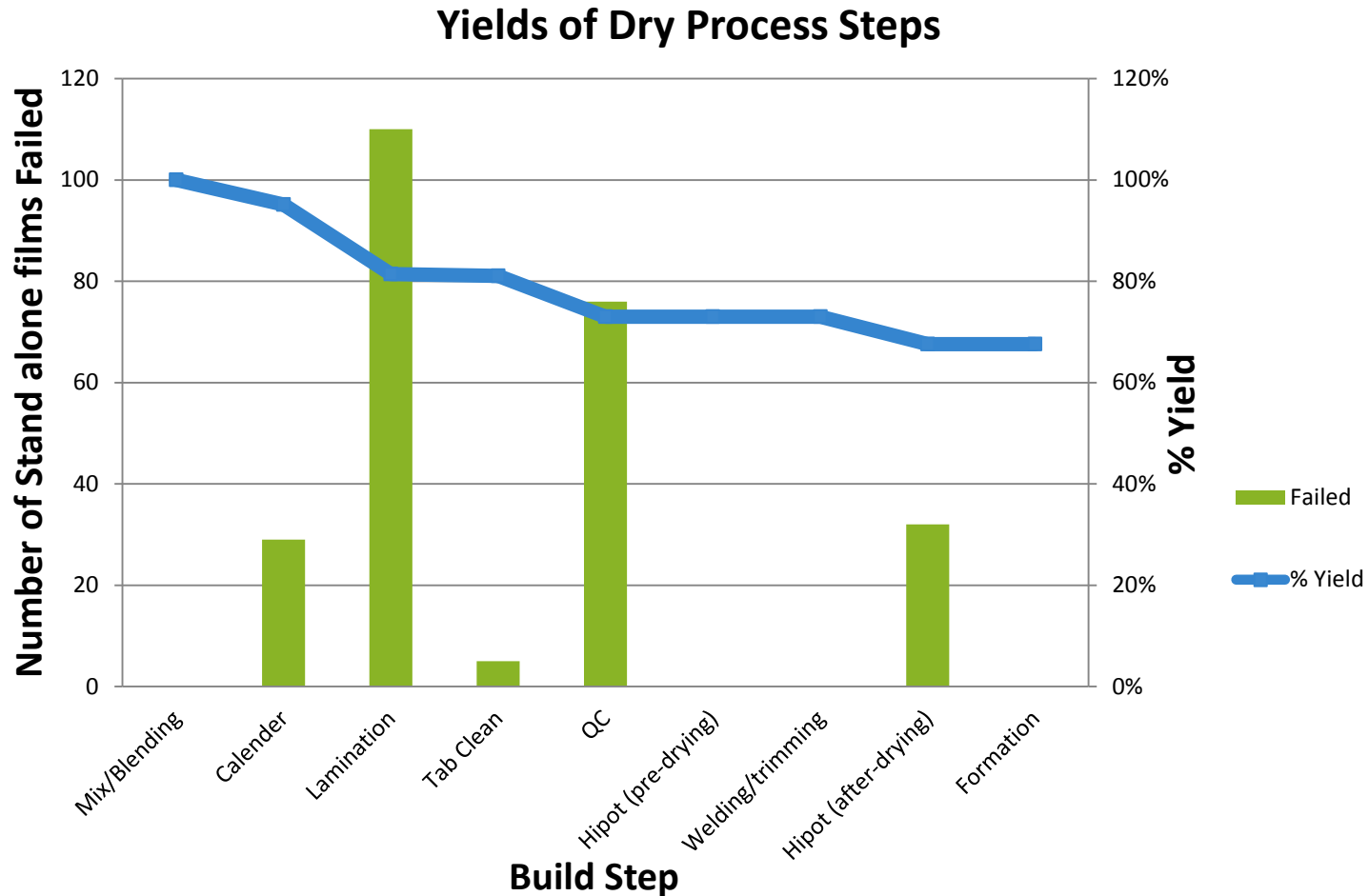


19 cells built. Met project milestone with cell delivery to ANL on 3/31/14

Interim cell build - cathode yield data



Most defects were attributable to electrode tearing during calendaring, but were discovered at various points in fabrication.



Uniformity of powder feed and loading is key to improving cathode yield.

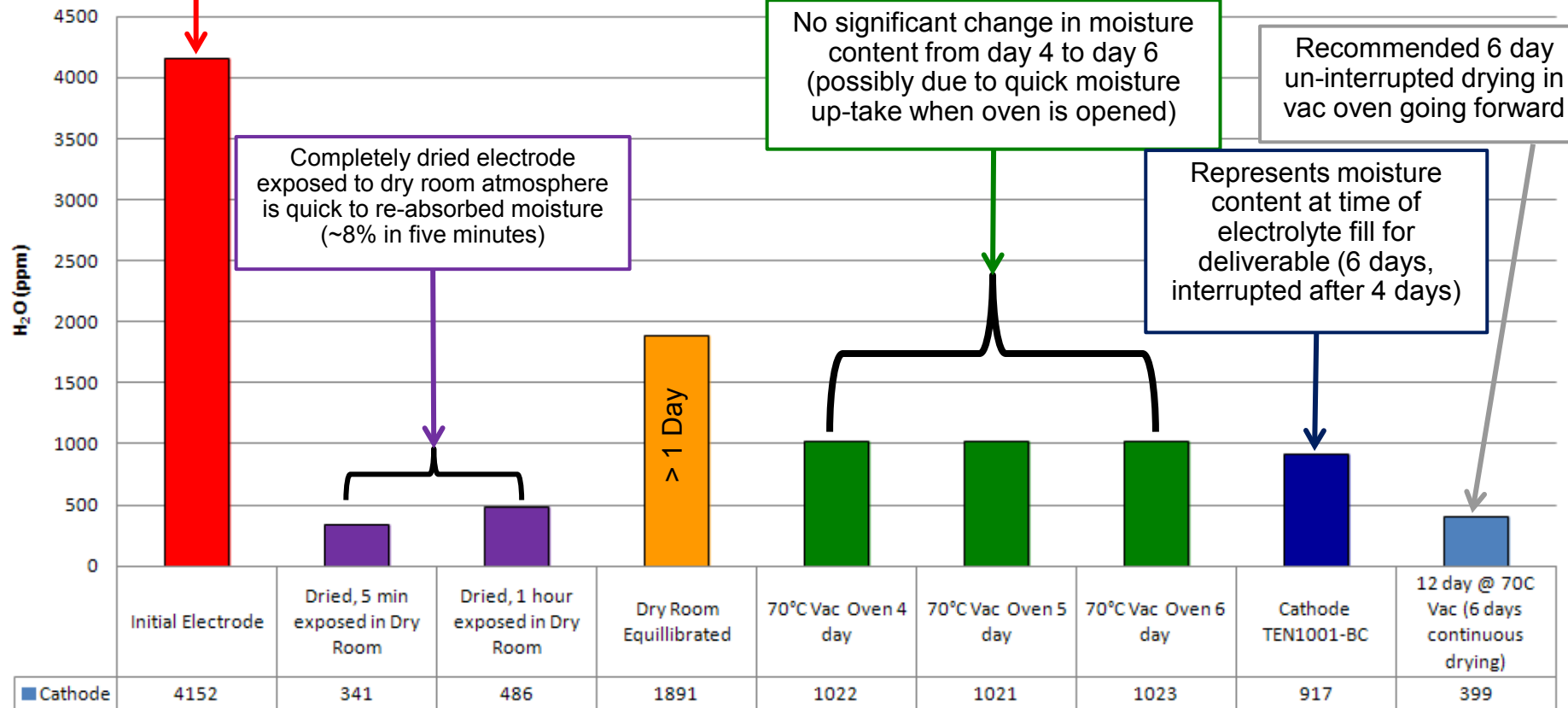
Residual moisture emerged as concern with dry process cathodes.



High initial moisture content arises from the hydrophilic nature of LMFP and high surface area activated carbon

Moisture target of < 1000 ppm was met for this deliverable; however, extended drying is required

Moisture Analysis of Dry Process Cathode



Response to Reviewer Comments



Reviewer 2: ...the approach is questionable in terms of speed of fabrication compared to the coating process. Therefore the relative cost savings is questionable.

Cost reduction is attained through eliminating capex and opex needed to operate 30+m long drying train with NMP drying and recovery. For the dry process, reducing the number of steps is more significant than simply increasing speed, and will be the focus of future work.

Reviewer 3: The cathode adhesion/cohesion, loading level, and porosity are far from optimum level for decent rate and cycle life. No controls were provided for comparison.

The data in slides 7 and 13 show acceptable life and rate from dry process cathode for EV application. The target loading and porosity are consistent with NCA and NMC cathodes optimized by Navitas for high energy cell designs. These cathodes have loadings of 4.5 – 5 mAh/cm² and porosity 27-30%, with >80% capacity retention at 1C (full cell).

Collaborations



- No relationship or tech transfer to Wanxiang/A123.
- Navitas is collaborating with process equipment OEM on anode Advanced Drying Process
- Dow Energy Materials has been non-funded collaborator in providing samples and guidance for LMFP cathode material
- DuPont R&D staff have provided samples and consulting on PTFE binder choice and process control.

Remaining Barriers and Challenges



- Increase cathode width for EV footprint - limited by calender pressure.
- Reduce number of calender passes and improve cathode yield.
- Reduce dry anode initial capacity loss from 30% to <15%.
- Capitalize roll-to-roll implementation of anode advance drying process.

Future work

Cathode

Qualify alternative processing additive to mitigate moisture retention and increase active material content.

Modify calender to reduce number of passes and increase width of free-standing cathode films.

Anode

Reformulate dry anode to reduce initial capacity loss

Down-select and scale-up anode process for final deliverable EV cell

Initial demonstration of full cell combining low-cost process anode and cathode.

Summary slide



- The dry electrode process innovation in this proposal will provide the ability to coat thick *and* fast, while eliminating solvents and saving energy.
- The projected readiness level is TRL 7 for the cathodes upon completion of the program, with confidence that the development path will leverage Zn-air or ultracapacitor production technology.
- Sound mechanistic understanding of the cathode process combined with Navitas' understanding of anode binder chemistry/electrochemistry will enable a new binder and dry process for anode.

Acknowledgments



- Cathode: Bob Sosik
- Anode: Pu Zhang
- Characterization: Danny King
- Cell Build: Brian Glomski

- Brian Cunningham and Ralph Nine, Department of Energy