

U.S. DEPARTMENT OF ENERGY'S (DOE)  
VEHICLE TECHNOLOGIES OFFICE (VTO)  
2020 ANNUAL MERIT REVIEW (AMR)



## BATTERY DESIGN FOR RECYCLE

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## RECELL CENTER FOR ADVANCED BATTERY RECYCLING



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Oak Ridge National Laboratory

bat467  
Virtual Poster  
June 1-4, 2020

# PROJECT OVERVIEW

## Timeline

- Project start: October 2018
- Project end: September 2021
- Percent complete: ~50%

## Budget

FY19	\$4,615k
FY20	\$5,150k

## Barriers

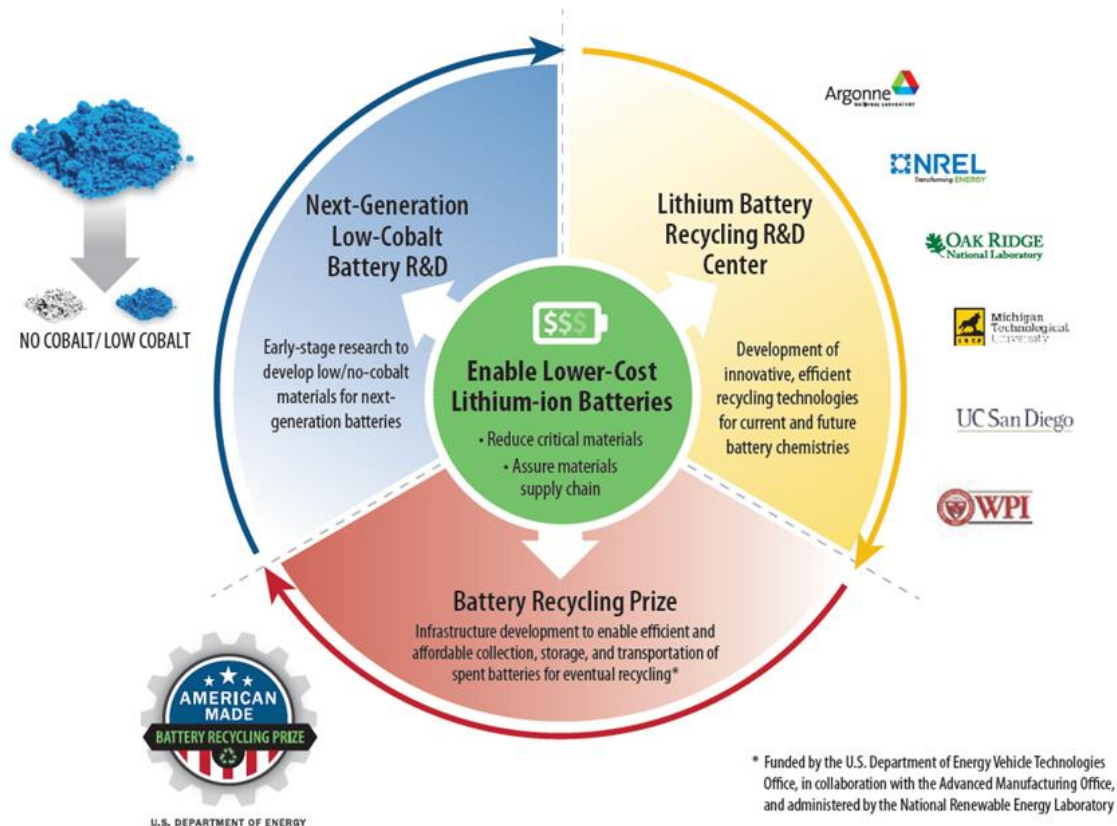
- Recycling and Sustainability
  - Cost to recycle is currently 5-15% of battery cost
  - Material shortage (Li, Co, and Ni)
  - Varying chemistries result in variable backend value

## Partners

- Argonne National Laboratory
- National Renewable Energy Laboratory
- Oak Ridge National Laboratory
- University of California, San Diego
- Worcester Polytechnic Institute
- Michigan Technological University

# RELEVANCE

- Lower cost of batteries
- Enable lower environmental impacts
- Increase our country's energy security



# APPROACH

Year 1 – Bench scale testing:  
Powder-to-Cell



Year 2 – Start to scale up  
unit operations

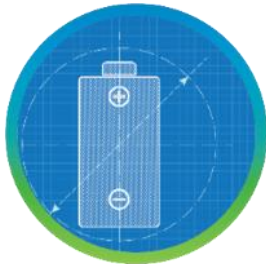


Year 3 – Finish scale up and  
show cell-to-cell recycling



**DIRECT  
CATHODE  
RECYCLING**

**OTHER  
MATERIAL  
RECOVERY**



**DESIGN  
FOR  
RECYCLING**

**MODELING  
AND  
ANALYSIS**



*ReCell does not include battery dismantling, transportation, or 2<sup>nd</sup> use*

# PROGRAM MILESTONES

- FY19 Q1 **Complete** Establish the battery recycling center's mission and include its targets and goals
- FY19 Q2 **Complete** Provide an initial progress report on roll-to-roll relithiation
- FY19 Q3 **Complete** Provide an initial progress report on design for recycle initiative
- FY19 Q4 **Complete** Establish the ReCell Center's Battery Recycling Laboratory and Scale-up Facility
- 
- FY20 Q1 **Complete** Electron Backscatter Diffraction data comparison of various chemically delithiated NMC-111 versus pristine NMC-111
- FY20 Q2 **Complete** All five relithiation processes added to EverBatt at lab scale and production scale
- FY20 Q3 **Ongoing** Down-select solvent(s) to separate black mass from current collector and optimize the process conditions to achieve >90% recovery of black mass
- FY20 Q4 **Ongoing** Demonstrate recovery of anode and cathode powders using the new pilot scale froth column

*Each Individual project has its own milestones that are not listed here.*

# DESIGN FOR RECYCLE

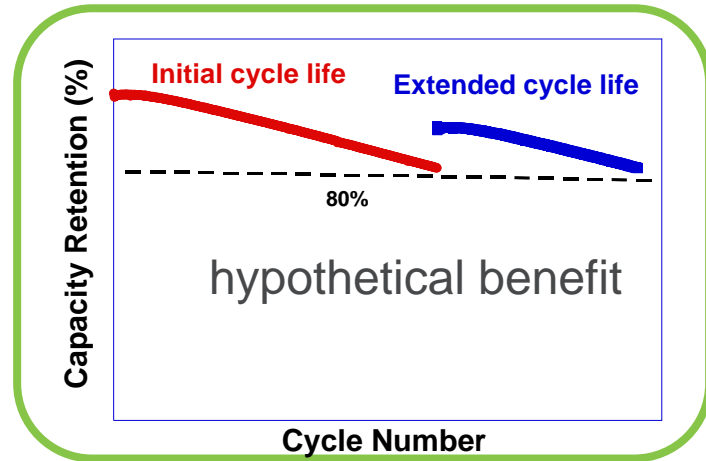
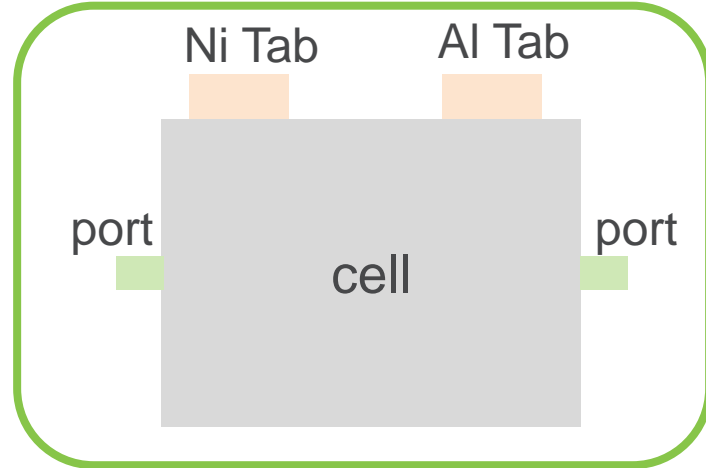
# CELL DESIGN

## Project Goal:

- Create cell designs to enable rejuvenation of a spent cell that trade minimal loss in energy-density performance for the ability to use cheaper, new recycling processes at end of life
- Relithiate spent cathodes to extend battery cycle life and reduce the number of batteries to be recycled

## Project Description:

- Develop new cell designs with ports that allow us to flush off some SEI components and reduce cell polarization
- Identify optimal rejuvenation conditions, including solvents, flushing times and period
- Demonstrate extended cycle life in rejuvenated cells



# FABRICATED BASELINE ELECTRODES AND VALIDATED THEIR ELECTROCHEMICAL PERFORMANCE

## Anode: LN3174-63-1 (single-sided)

91.83 wt% Superior Graphite SLC1520P  
2 wt% Timcal C-45  
6 wt% Kureha 9300 PVDF Binder  
0.17wt% Oxalic Acid

ReCell, 18650 cell build with cathode [LN3174-64-3&65-3]  
Prod: SLC1520P, Lot#: 022626-376-551

"SS" = single sided -> **CALENDERED**

Cu Foil Thickness: 10  $\mu$ m

Total Electrode Thickness: 98  $\mu$ m (SS)

Coating Thickness: 88  $\mu$ m (SS)

Porosity: 34.5 %

Total SS Coating Loading: 12.49 mg/cm<sup>2</sup>

Total SS Coating Density: 1.42 g/cm<sup>3</sup>

Estimated Areal Capacity: 3.78 mAh/cm<sup>2</sup>

[Based on rev. C/10 of 330 mAh/g for 0.005 to 1.5 V vs. Li]

Made by CAMP Facility

## Cathode: LN3174-65-3

(single-sided)

90 wt% Targray NMC622

5 wt% Timcal C-45

5 wt% Solvay 5130 PVDF Binder

ReCell, 18650 cell build with anode [LN3174-62-4&63-1]

Prod: Targray SNMC03006, Lot#: TBD

"SS" = single sided -> **CALENDERED**

Al Foil Thickness: 20  $\mu$ m

Total Electrode Thickness: 95  $\mu$ m (SS)

SS Coating Thickness: 75  $\mu$ m (SS)

Porosity: 33.5 %

Total SS Coating Loading: 20.43 mg/cm<sup>2</sup>

Total SS Coating Density: 2.72 g/cm<sup>3</sup>

Estimated SS Areal Capacity: 3.11(3.27) mAh/cm<sup>2</sup>

[Based on rev. C/10 of 169(178) mAh/g for 3.0 to 4.2(4.3) V vs. Li]

Made by CAMP Facility

## Anode: LN3174-62-4 & 63-1 (double-sided)

91.83 wt% Superior Graphite SLC1520P  
2 wt% Timcal C-45  
6 wt% Kureha 9300 PVDF Binder  
0.17wt% Oxalic Acid

ReCell, 18650 cell build with cathode [LN3174-64-3&65-3]  
Prod: SLC1520P, Lot#: 022626-376-551

"SS" = single sided, "DS" = double sided -> **CALENDERED**

Cu Foil Thickness: 10  $\mu$ m

Total Electrode Thickness: 180  $\mu$ m (DS)

SS Coating Thickness: 85  $\mu$ m (SS)

Porosity: 32.2 %

Total SS Coating Loading: 12.49 mg/cm<sup>2</sup>

Total SS Coating Density: 1.47 g/cm<sup>3</sup>

Estimated SS Areal Capacity: 3.78 mAh/cm<sup>2</sup>

[Based on rev. C/10 of 330 mAh/g for 0.005 to 1.5 V vs. Li]

Made by CAMP Facility

## Cathode: LN3174-64-3 & 65-3

(double-sided)

90 wt% Targray NMC622

5 wt% Timcal C-45

5 wt% Solvay 5130 PVDF Binder

ReCell, 18650 cell build with anode [LN3174-62-4&63-1]

Prod: Targray SNMC03006, Lot#: TBD

"SS" = single sided, "DS" = double sided -> **CALENDERED**

Al Foil Thickness: 20  $\mu$ m

Total Electrode Thickness: 165  $\mu$ m (DS)

SS Coating Thickness: 73  $\mu$ m (SS)

Porosity: 31.2 %

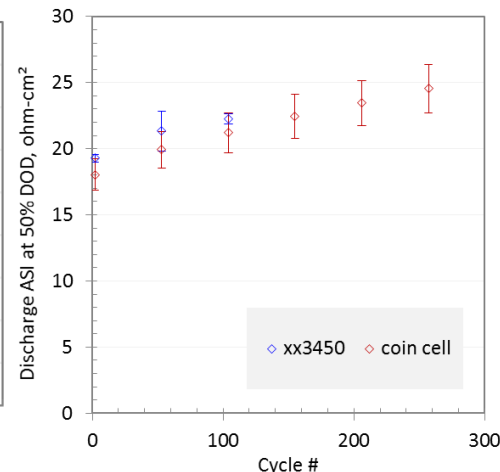
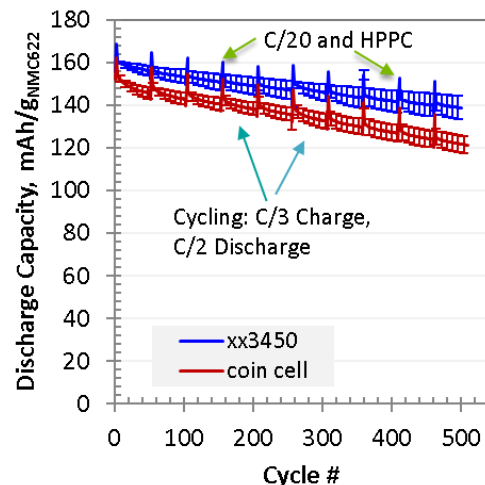
Total SS Coating Loading: 20.43 mg/cm<sup>2</sup>

Total SS Coating Density: 2.82 g/cm<sup>3</sup>

Estimated SS Areal Capacity: 3.11(3.27) mAh/cm<sup>2</sup>

[Based on rev. C/10 of 169(178) mAh/g for 3.0 to 4.2(4.3) V vs. Li]

Made by CAMP Facility



- Validated excellent electrochemical performance from the baseline electrodes
- Fabricated cylindrical and pouch cells
- Created spent cells/electrodes for rejuvenation



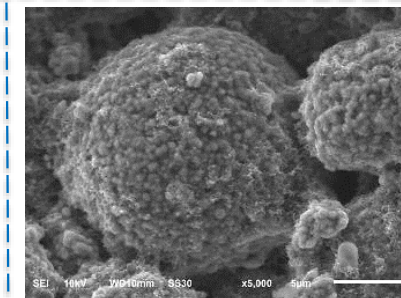
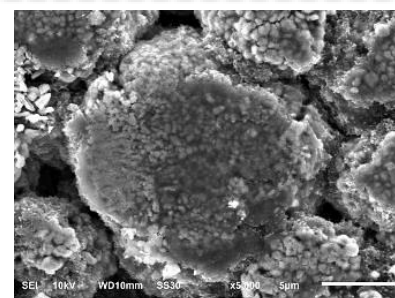
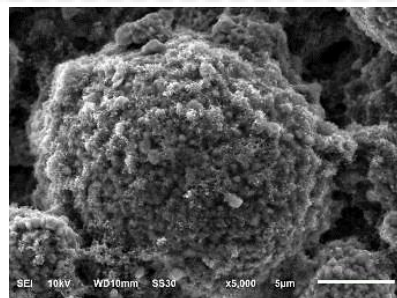
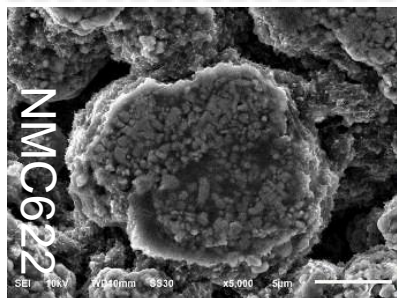
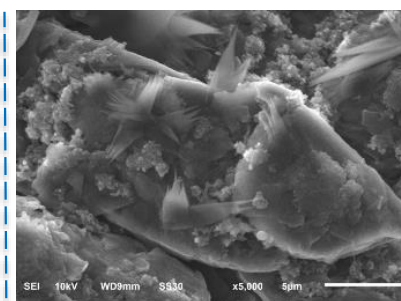
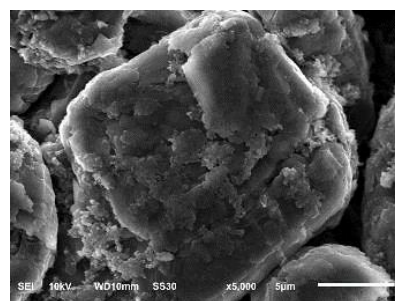
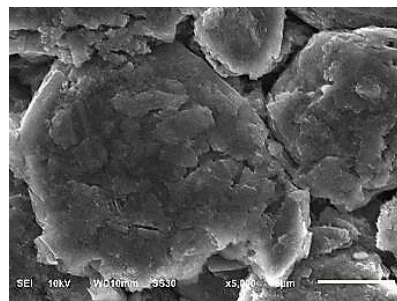
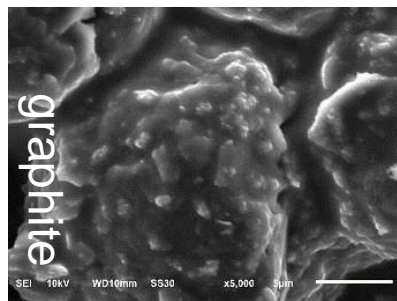
# ELECTRODE RINSING REMOVED THE SURFACE LAYER FROM SPENT GRAPHITE ANODE BUT HAD LITTLE EFFECT ON THE SPENT CATHODE

Unwashed

DMC

IPA

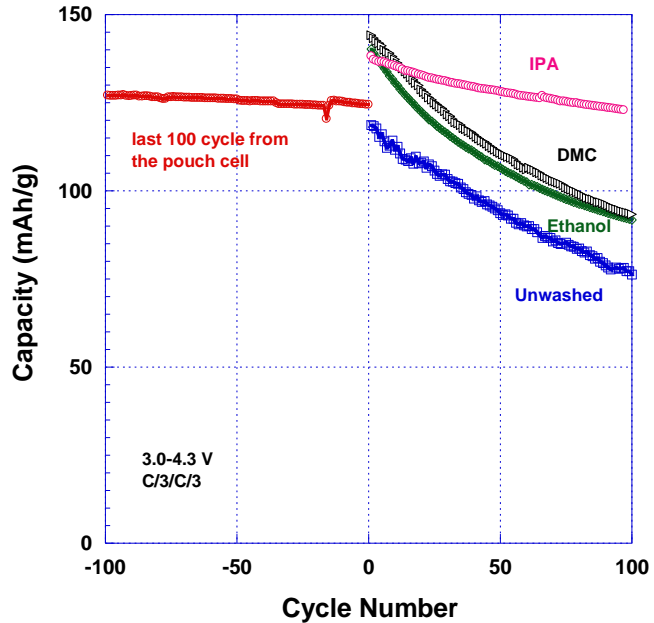
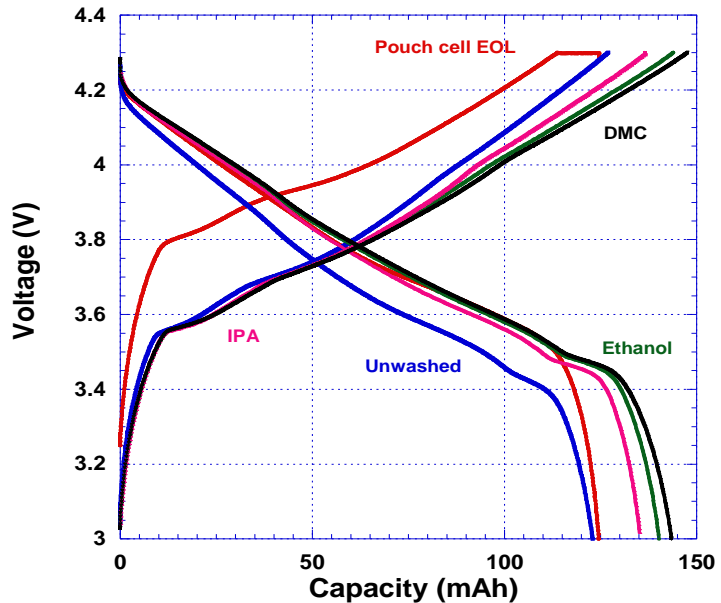
EtOH



XPS results indicated  $\text{Li}_2\text{CO}_3$  was removed after rinsing.

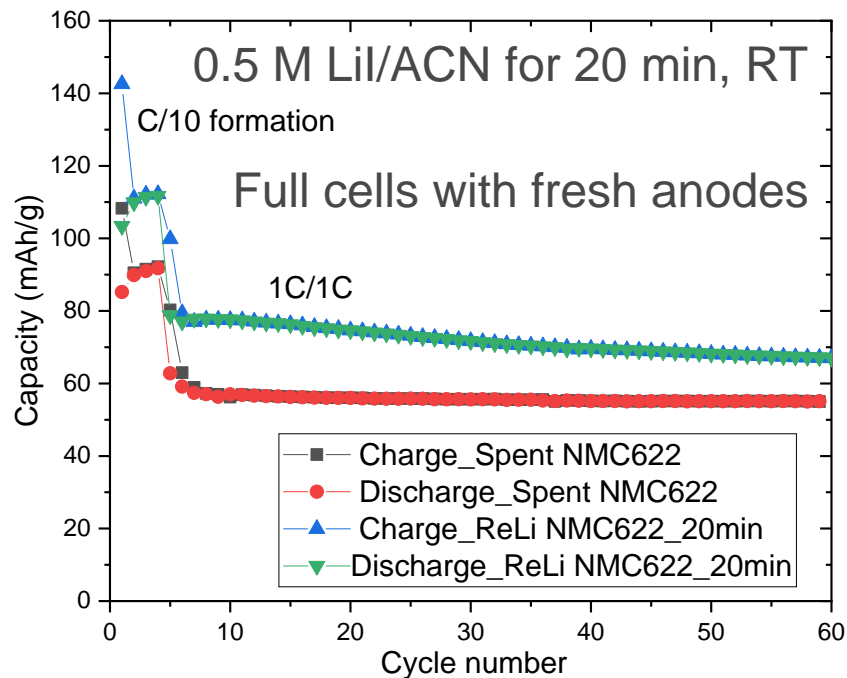
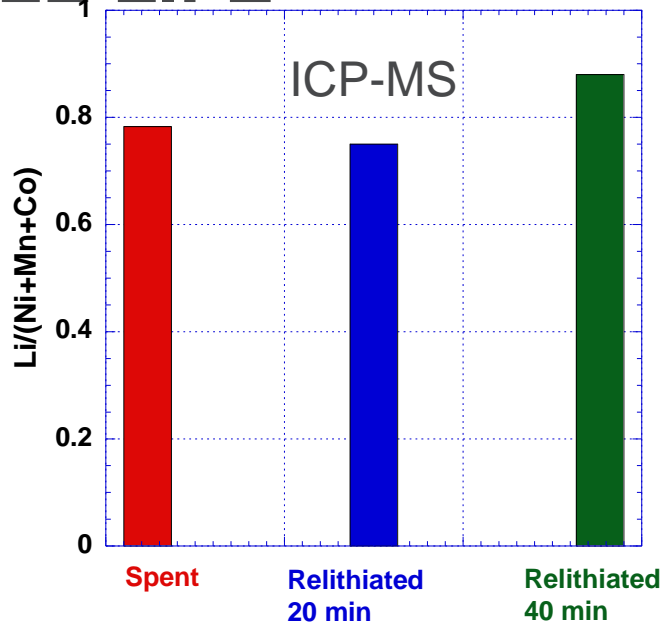
Rinsing period: 5 min

# INITIAL RESULTS FROM RINSING SPENT ELECTRODES SHOWED CAPACITY RECOVERY



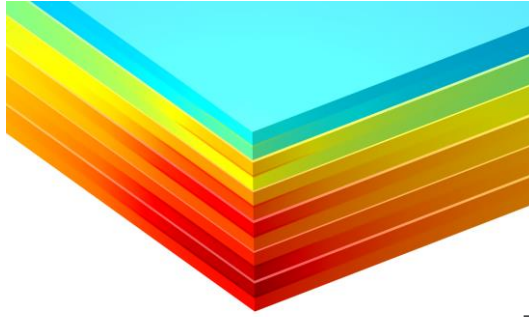
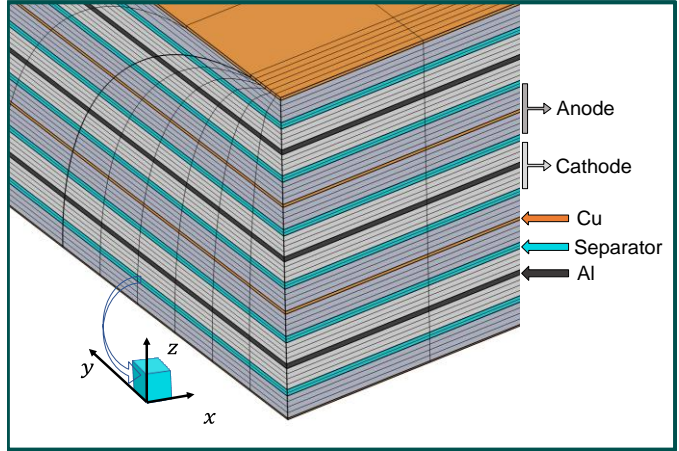
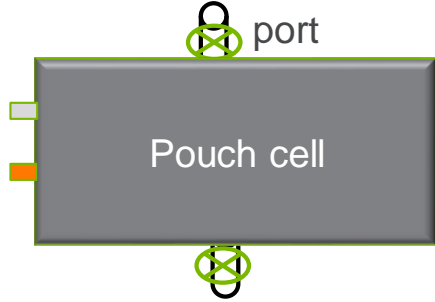
- Rinsing the spent electrode improved the capacity in the re-assembled cells albeit subsequent fast capacity degradation.
- The rinsed solution from the spent electrodes mainly consisted of  $\text{POF}_3$ ,  $\text{C}_5\text{H}_{10}\text{O}_3$ ,  $\text{C}_7\text{H}_{14}\text{O}_5$  and  $\text{C}_8\text{H}_{14}\text{O}_6$  esters while  $\text{C}_2\text{H}_6\text{O}$  was also found from the anode solution.

# RINSING AND RELITHIATING SPENT NMC622 SHOWED RECOVERY IN CAPACITY AND EXTENDED CYCLE LIFE



- Li content in spent NMC622 increased after relithiation for 40 min
- Relithiated electrodes showed higher capacity.

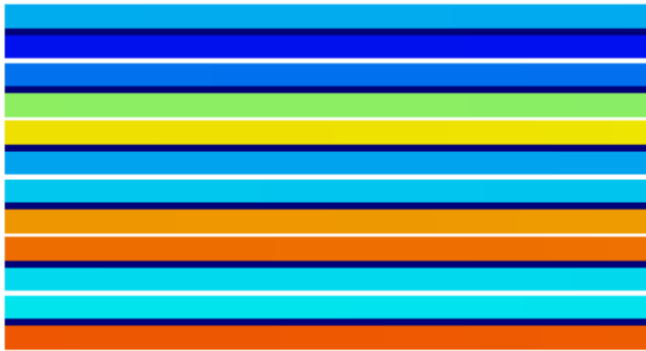
# HIGHER PRESSURE AND HIGH FLUX AT THE COMPONENTS WITH HIGHER PERMEABILITY



0 1 2 3 4 5 6  $\times 10^5$  Pressure, Pa

- NMC622
- SLC1520P
- Celgard separator

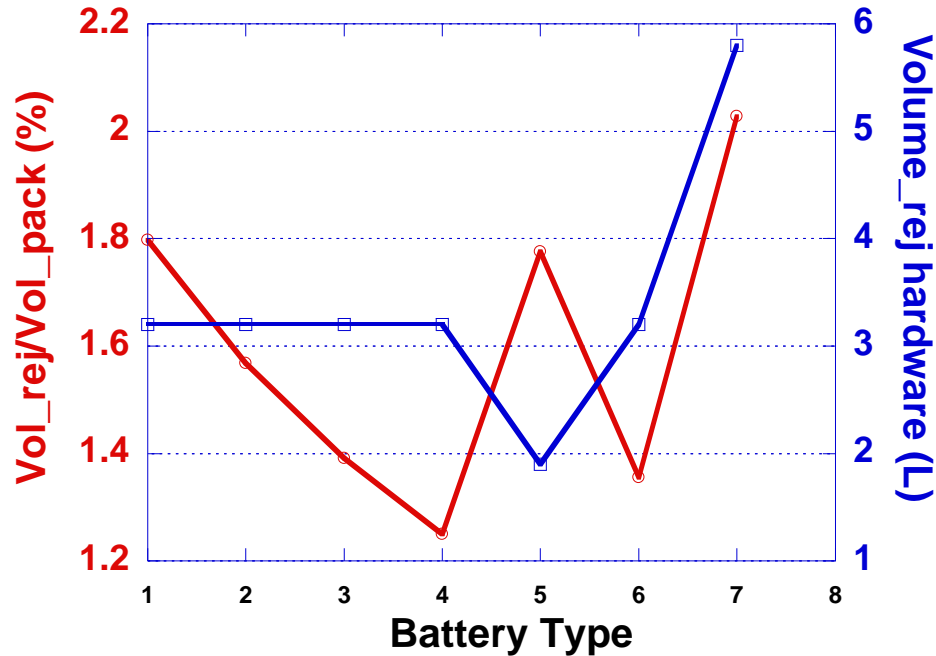
Flow velocity through cell mid-section



5 10 15 20 25 30 35 40 45 50  $\times 10^{-5}$

Flow rate m/s

# REJUVENATION HARDWARE WOULD COMPROMISE ENERGY DENSITY BY 1-2%



- Estimation using BatPac
  - Assume: 2 tubes per cell, each 30 cm long & 0.4 cm OD (~4 mL each)
  - Assume: 2 valves per module (30 cells per module), ~60 mL each
  - Assume: 2 ganged tubing ports per module, ~15 mL each
  - Implied total of 400 mL per module if no wasted volume
  - Volume of the hardware for rejuvenation is 1.9 to 5.8 L.
- The additional rejuvenation hardware would increase the pack volume by 1-2%.
- Courtesy of Andy Jansen

# DESIGN FOR RECYCLE – ACCOMPLISHMENTS AND RESULTS

- Created new cell design for cell rejuvenation
- Created spent cells and electrodes with NMC622 and SLC1520 P graphite
- Demonstrated capacity recovery after rinsing the spent electrodes in coin cells
- Demonstrated capacity recovery from relithiating the spent NMC622 cathodes
- Simulated flow rate and pressure distribution in pouch cells during electrode rinsing
- Estimated penalty in energy density with the new cell design using BatPac

For more information please see [www.recellcenter.org](http://www.recellcenter.org), where our Quarterly Reports are posted.

# COLLABORATION AND ACKNOWLEDGEMENTS

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Jiuling Yu (NREL)  
Ruiting Zhan (MTU)





# RECELL RECYCLING TOWN HALL

## FRIDAY, JUNE 5, 2020 FROM 1:00 TO 3:00 (CENTRAL)

To continue the discussion the ReCell team will hold an interactive town hall meeting. Please join us at the BlueJeans session shown below and ask questions through Slido



Take a picture of  
this slide

For Information  
about ReCell



### *BlueJeans Meeting Access information*

Computer

<https://bluejeans.com/749203749/9534?src=htmlEmail>

Phone

(866) 226-4650

Meeting ID: 749 203 749

### *Slido Q/A website*

Computer or Smart Phone

[www.Slido.com](http://www.Slido.com)

Event Code

“recell”