Physical Sciences Inc.

Construction of High Energy Density Batteries (DOE Phase II SBIR)

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10 April 2017

Project ID # ES288

Acknowledgement: This material is based upon work supported by the Department of Energy under Award No. DE-SC0011877.

Overview

Timeline

- Project Start (Phase I): June 9, 2014
- Project Start (Phase II): July 28, 2015
- Project End: July 27, 2017
- Percent Complete: 82%

Budget

- FY15-16 = $1000K (DOE)

Barriers Addressed

- **Performance:** The proposed technologies reduce the inactive mass in the cell increasing cell energy density.
- **Cost:** The PSI coating technology reduces cost by enabling increased active material loadings and operating voltages, thus reducing the amount of cathode material and solvent required during cell production.

Project Partners

- Argonne National Laboratory
- SKC Powertech, Inc.

Relevance and Project Objectives

Overall Project Objective: Construct cells offering a 25% increase in cell energy density over the state of the art.

- Demonstrate the PSI high active (HA) coating technique allows production of high performance cathode electrodes with >98% active material that:
  - Deliver equivalent electrochemical performance to conventional electrodes.
  - Require reduced amount of solvent during electrode production.
  - Enable stable, extended high voltage operation.
- Produce 3+Ah cells using HA cathode material and demonstrate equivalent cycling performance to cells with a conventional electrode formulation.
- Demonstrate a novel composite film with <30% the mass of conventional Cu foil can be used as the anode current collector in a Li-ion cell.

Path to realizing increase in cell energy density:

- HA contribution: 3-5% increase in gravimetric (4-6% increase in volumetric)
- HA + high voltage (HV): 10-14% increase in gravimetric
- HA + HV + higher loading (HL): 13-17% increase in gravimetric (>22% in volumetric)
- HA + HV + HL + anode composite current collector: 25-29% increase in gravimetric

Projected increase in baseline cell energy density from ~197Wh/kg to 247Wh/kg on incorporating all technologies.

Milestones

Date	Milestones	Status
January 2016	Cathode Electrode Scale-Up: Kg- scale coating of cathode material/ electrode production	Complete
February 2016	Pouch Cell Construction: Production of 3+Ah cells with PSI coated material	Complete
June 2016	Anode Current Collector Scale-up: Continuous production and demonstration in lithium ion cells.	Complete
June 2016	Cycling and Rate Testing: Confirmation of required pouch cell performance and cycle life.	Complete
July 2016	Pouch Cell Construction: Production of high voltage 3+Ah cells with PSI coated material	Complete
June 2017 (Revised)	Pouch Cell Construction: Production of 3+Ah cells with PSI cathode and anode current collector technologies. (Efficient cycling of smaller cells completed.)	On track to meet revised date.
June 2017 (Revised)	Cycling and Rate Testing: Confirmation of optimized pouch cell performance and cycle life.	On track. Cells being constructed in coming month.

Program Approach/Strategy

Phase II efforts are focused on producing multi-amp hour cells.

- **Focus of Year 2 (July 2016 through July 2017):**
  - Novel Composite Anode Current Collector
    - Demonstrate performance of continuously produced films and optimize conductivity.
    - Confirm performance in multi-layer pouch cells.
    - Evaluate performance in 2+Ah sized pouch cells.
  - Cathode Coating/Electrode Scale-Up
    - Apply PSI HA coating to kg levels of NCM-622 cathode material to support multiple cell construction efforts.
    - Produce 3+Ah lithium ion cells designed for high voltage operation.
    - Evaluate performance of PSI and SKCP produced cells to demonstrate the required performance for use in electrode vehicles.
    - Work with scientists at ANL's CAMP facility to characterize the electrodes formed with multiple active materials using the PSI coating and electrode formulation techniques.

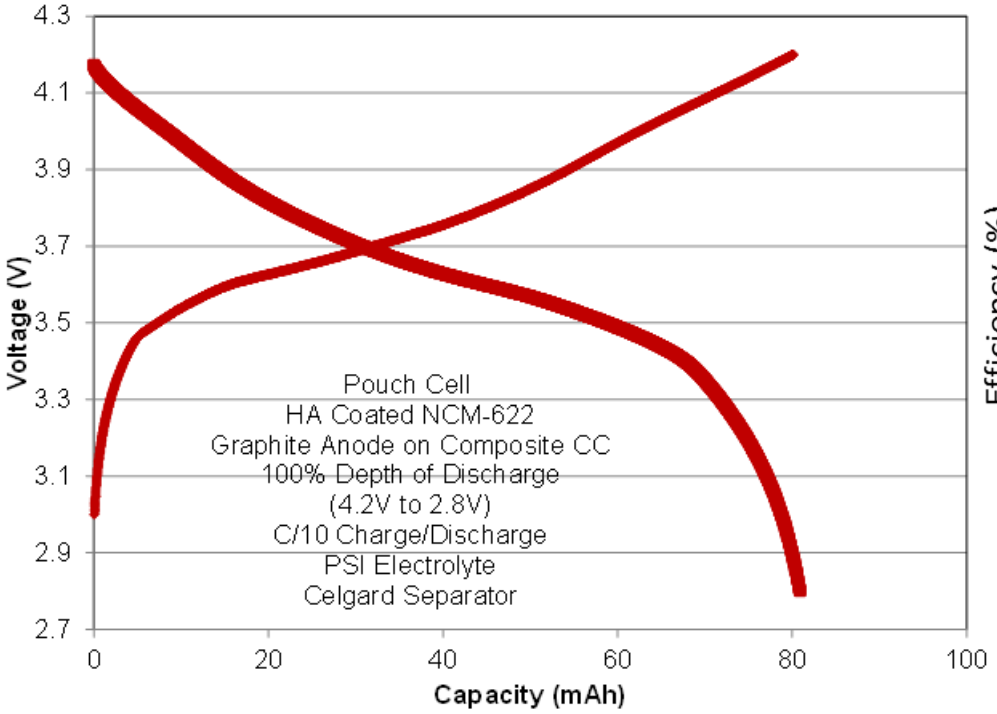
Continuous Composite Anode Current Collector Production

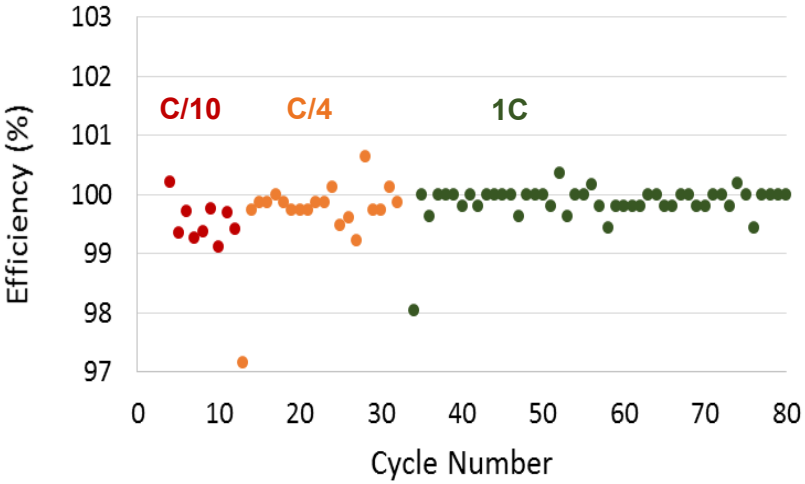
- Rolls of the composite current collector were prepared.
  - The mass was 2.55mg/cm^2 or <30% of the copper foil typically used at PSI.
  - The thickness can be tailored to 9-12μm comparable to traditional copper foil.
  - Double sided anodes were prepared on the composite using standard slurry casting, drying, and handling procedures.
  - Conductivity tests indicate increased electrical resistance of composite would reduce 1C discharge energy by <1.5%. Multi-layer testing underway to confirm.
- Electrodes were punched out using the standard electrode die.
- Pouch cells were built with up to 4 double sided anodes and PSI HA cathodes (330mAh).

Composite current collector roll and electrode for use in a pouch cell build.

HA Cathode and Composite Current Collector Pouch Cell Performance

- Extended cycling of full cells demonstrated robust electrical connection and no side-reactions with the composite substrate.
- Cycling tests were carried out at C/10, C/4, and 1C.
- The efficiency remains above 99% as the rate increases (C/10 -> 1C).
- The performance is maintained over 80+ cycles indicating minimal change in the electrical connection on cycling.





Charge/discharge voltage traces and cycling efficiency for pouch cell using the HA cathode and composite current collector.

Successful HA Coating of Commercial Cathode Materials

PSI has applied it's HA coating to a large number of commercially available cathode materials.

- Electrodes prepared with 98.5% HA material delivered equivalent performance to standard electrodes when normalizing for the active content.
  - Note the total HA mass is used to normalize the results.
- Solvent required during casting reduced by 50% or more.
- Electrode density increased due to reduced binder/conductor fractions.
  - >20% thickness reduction demonstrated increasing cell volumetric energy density.

Performance of HA electrodes formed using ANL supplied cathodes:

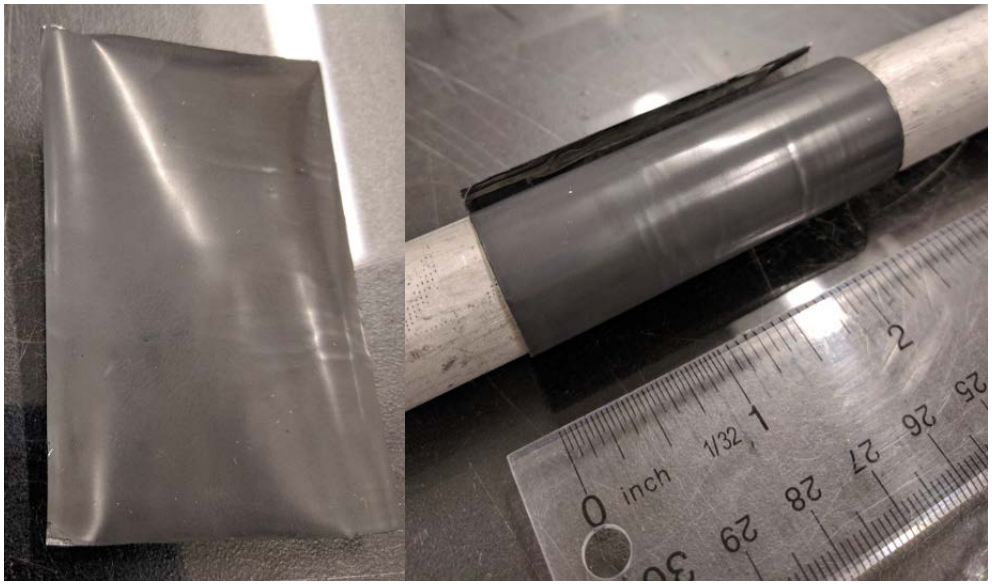
Cycling to normal voltage in half-cell, C/10 and **1C** discharge capacities.

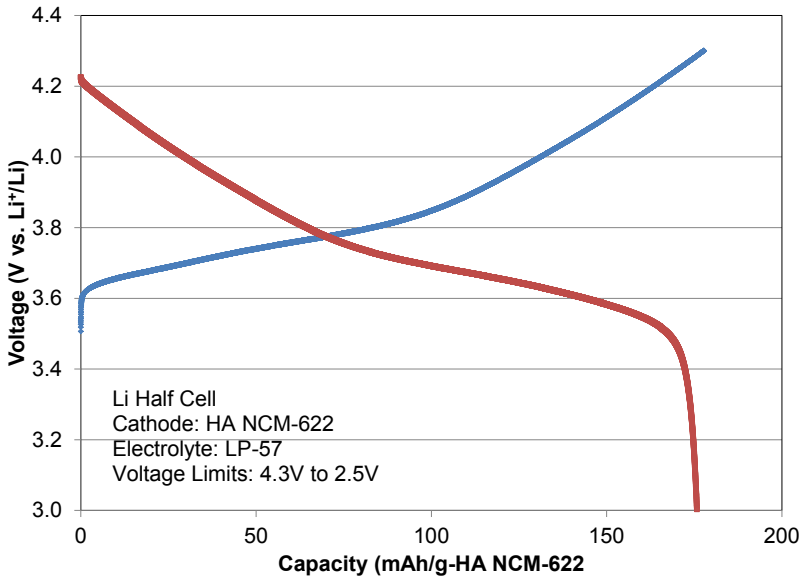
- $LiCoO_2$ (158/**154**mAh/g-HA), NCA (196/**184**mAh/g-HA), NCM-111 (157/**145**mAh/g-HA), NCM-523 (170/**159**mAh/g-HA), NCM-622 (182/**170**mAh/g-HA), NCM-811 (186/**177**mAh/g-HA).
- All values are consistent with those measured using uncoated cathode material and a traditional electrode formulation.

Production of Mechanically Robust High Loading Electrodes

HA coating enables production of high loading cathode electrodes with high active content (>98.5%) that are required for use in high energy cell designs.

- Electrodes were prepared with 98.75% HA NCM-622 at loadings >35mg/cm^2.
- The electrodes could be calendared, cut, punched, and rolled with no loss in material.
- The electrodes cycle efficiently with capacity values consistent with that measured on cycling thinner electrodes with this cathode material.
- No cracking, dusting, flaking, or change in the mechanical adhesion was noted after cycling of electrodes in a half-cell.



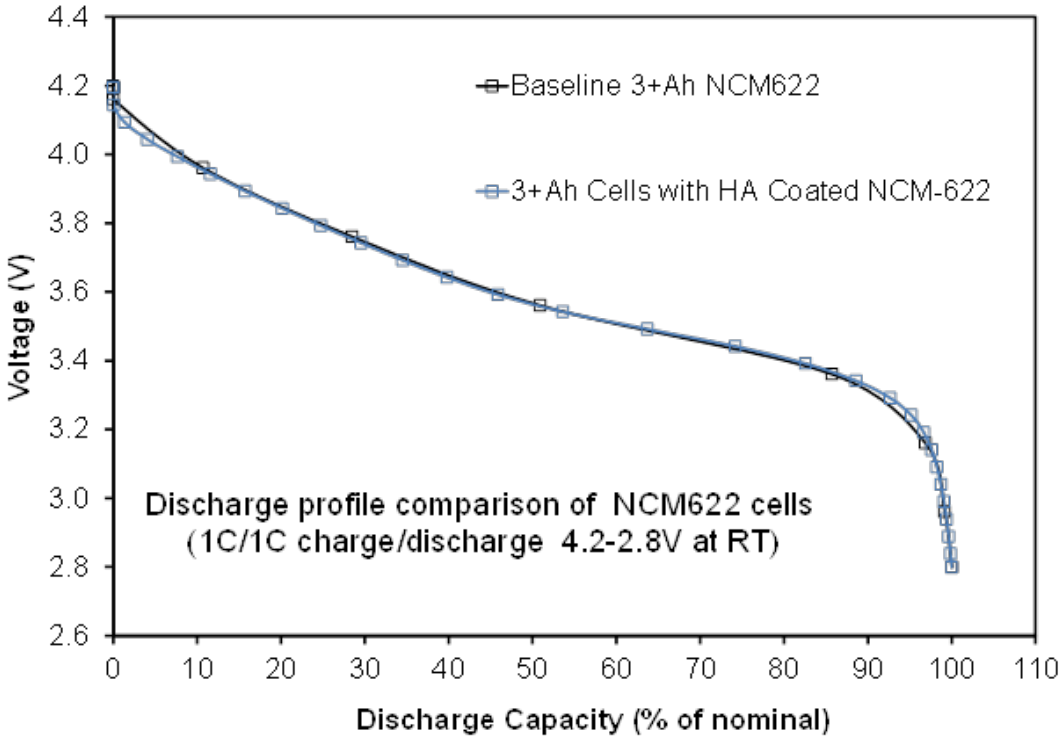


Flat and rolled HA NCM-622 electrode at >35mg/cm^2. Half-cell charge/discharge voltage traces.

Rate Performance of HA NCM-622 in 3+Ah Cells

Pouch cells were built with capacity >3Ah using the HA NCM-622 cathode material and electrode formulation.

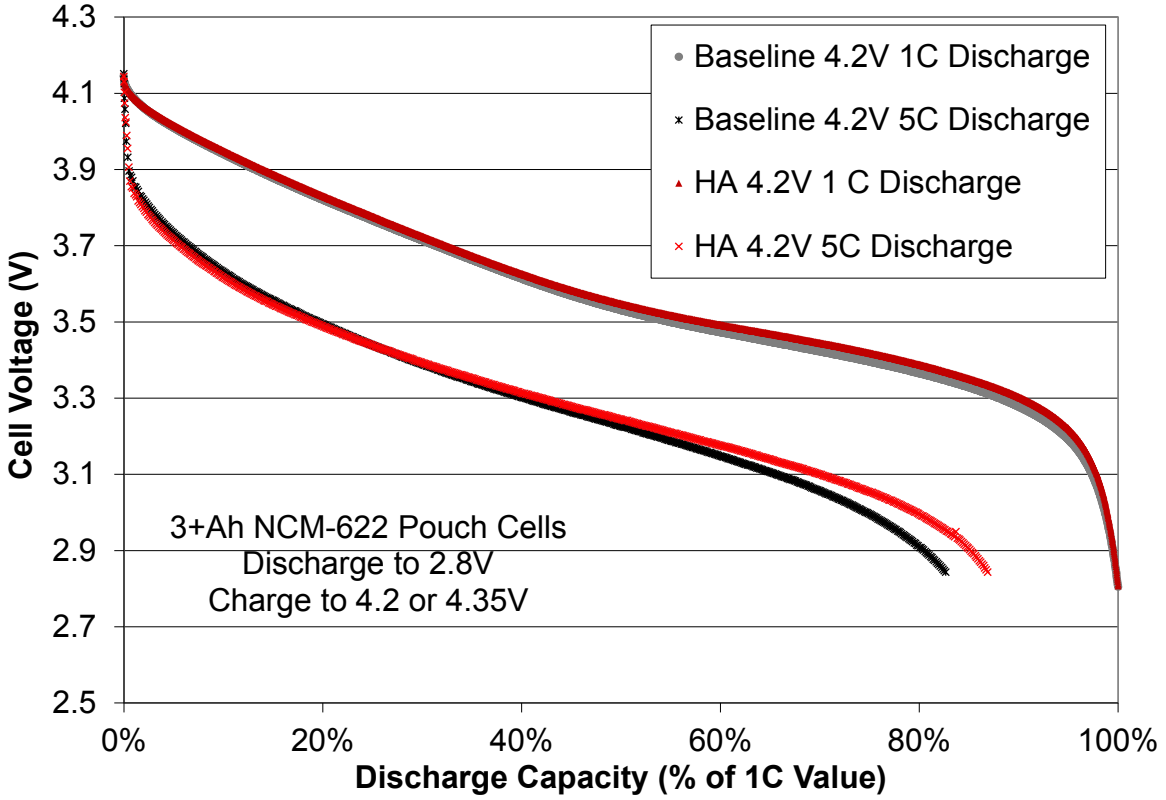
- The measured low rate capacity was 168-169mAh/g as targeted.
- The measured voltage profiles at 1C charge/discharge rates of the cells is consistent with that measured for the baseline cells (cells with standard cathode).



High Rate Performance of HA NCM-622 Cells

The HA cell on discharge at 5C delivers increased energy as compared to the baseline (the cell with the standard cathode).

- Energy densities of 268Wh/kg were achieved during 1C operation of a larger (5Ah) high power HA-cell designed for an internal PSI project.

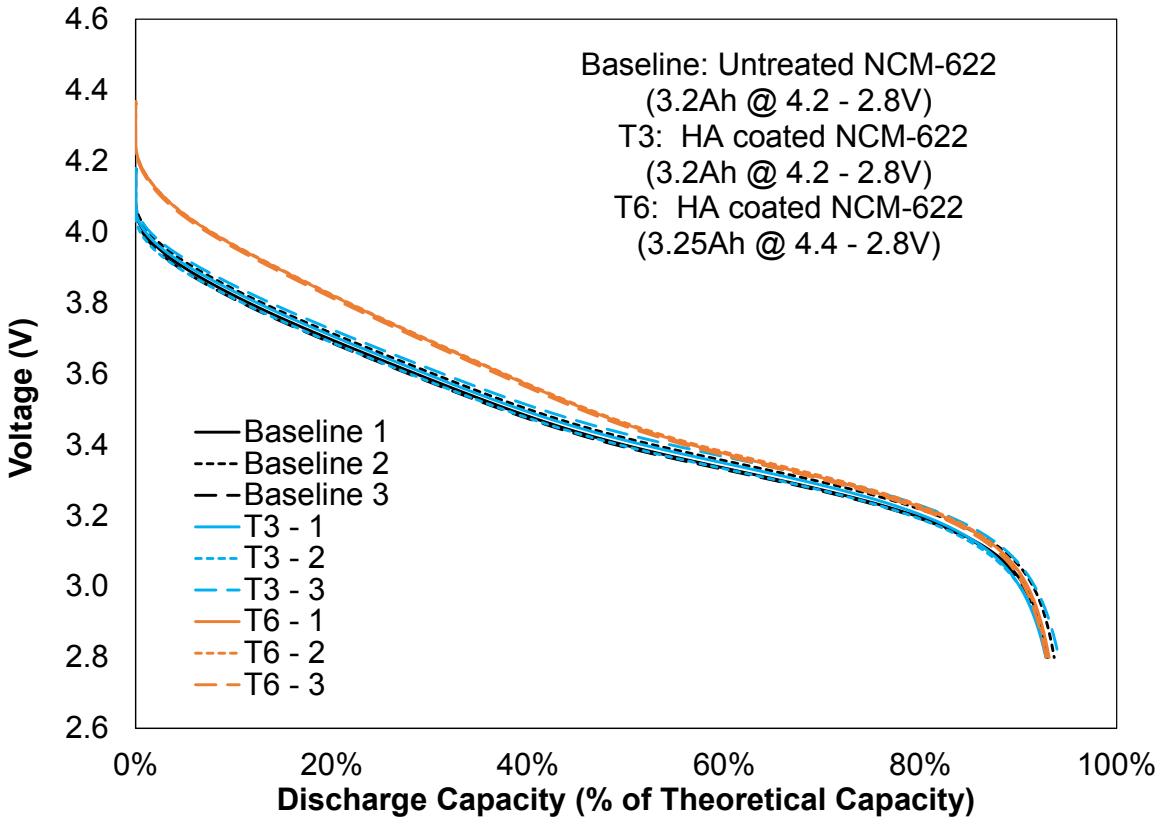


2C Rate Performance at Nominal and Elevated Voltages

Three ~3.2Ah cell versions were built and tested at various rates:

1. Baseline with a conventional NCM-622 cathode
2. HA NCM-622 with the loading optimized for operation at 4.2V
3. HA NCM-622 with the loading optimized for operation at 4.4V

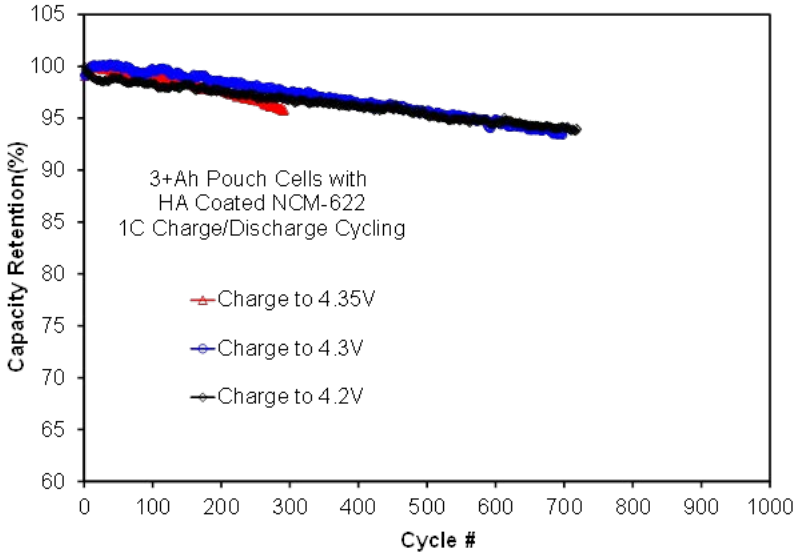
- The percentage of the capacity delivered at 2C for each pouch cell is equivalent.
- The energy delivered by the 4.4V HA cell is higher due to the higher average voltage.



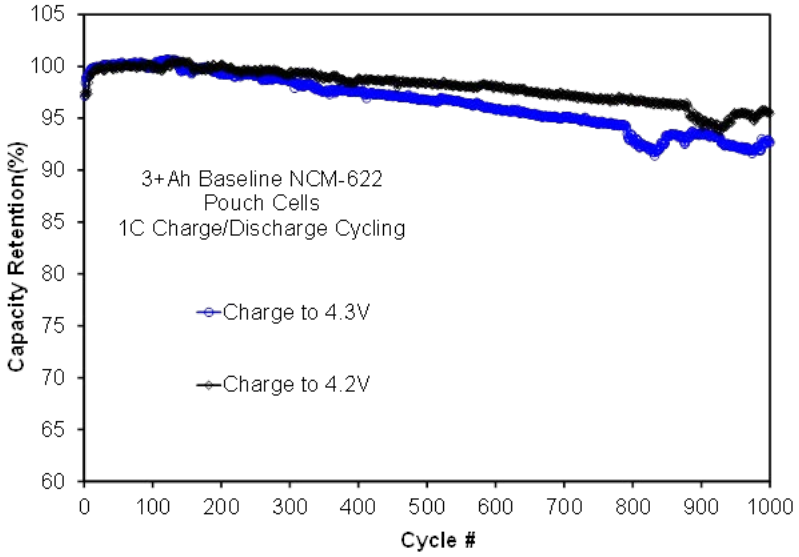
HA NCM-622 Pouch Cell Cycling at Various Maximum Voltages

1C full DOD charge/discharge cycling was performed to 3 different charge voltage levels (4.2, 4.3 and 4.35V) with minimal fade.

- The cycling performance is consistent with that for the baseline cells.
- The fade on cycling to 4.2 and 4.3V is ~1% per 100 cycles.
- The fade on cycling to 4.35V is <1.5% per 100 cycles.



Cells with HA NCM-622 Electrode

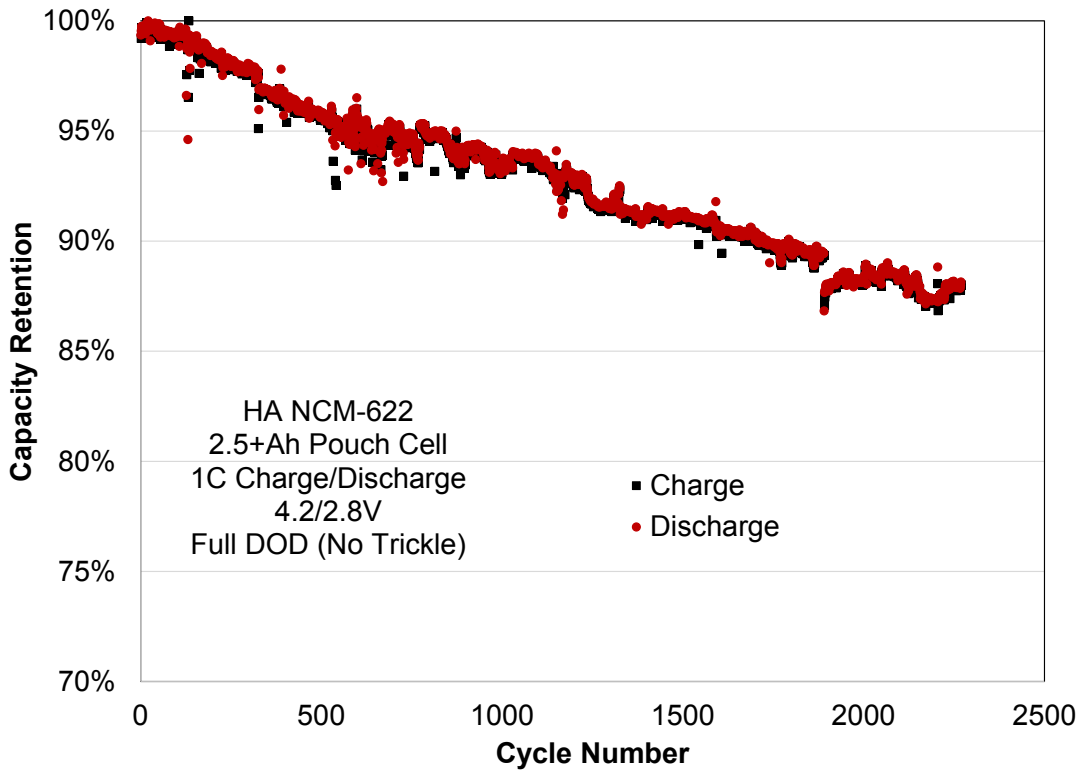


Cells with Baseline NCM-622 Electrode

Extended Cycling of HA NCM-622 3+Ah Pouch Cells

1C full DOD charge/discharge cycling was continued between 4.2 and 2.8V.

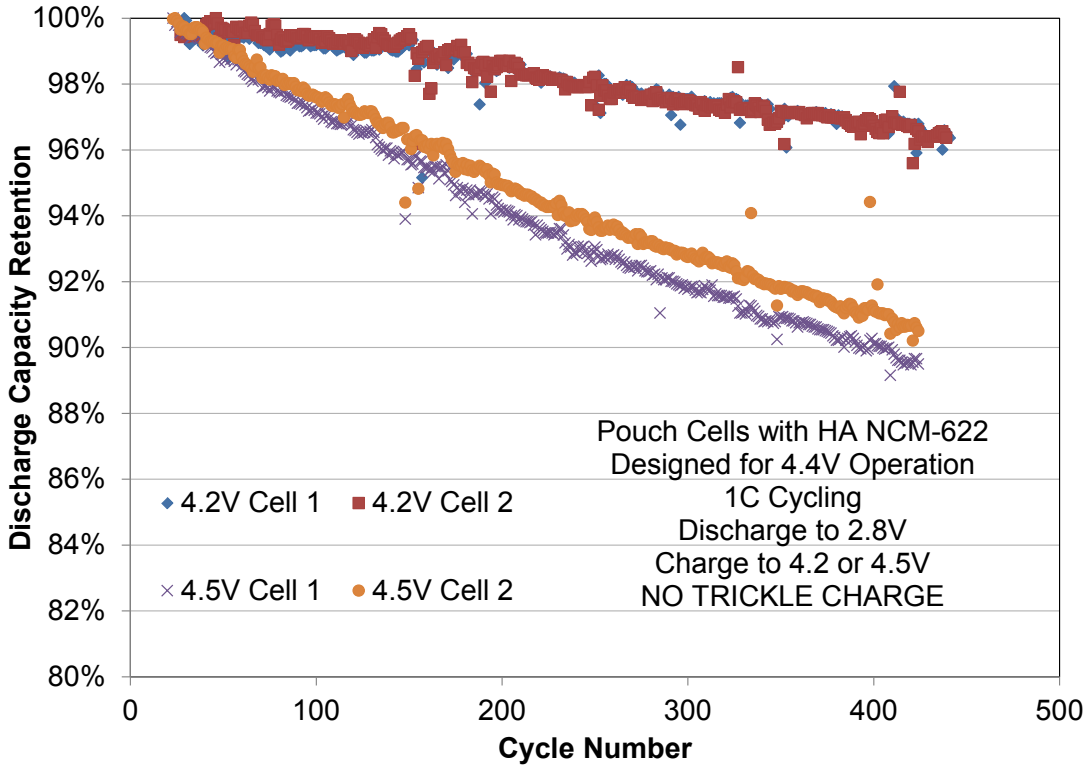
- The cell retained >90% of the initial capacity over 1700+ cycles.
- In total >2200 cycles were performed demonstrating the stability of the HA chemistry in the lithium ion environment.



Testing of HA NCM-622 Cells Designed for High Voltage Operation

Cells were built with the cathode loading targeted to allow operation at 4.4 and 4.5V.

- Tests were performed with the maximum voltage set to 4.2 and 4.5V first at C/2 and then 1C. No trickle charge of the cell was performed.
- The initial delivered capacity increases ~20% on increasing to 4.5V.
- The fade rate increases from ~1% per 100 cycles to 2.5% per 100 cycles. 800 cycles should be achievable with 80% of the initial charge.



Responses to Previous Year Reviewers' Comments

Reviewer Comment: PSI does not adequately describe how the 25% increase in energy density will be achieved. PSI must also better define the SOA.

Response: PSI has added information to illustrate the baseline cell used and the potential impact of each technology on the energy density.

Reviewer Comment: Insufficient information is provided regarding the composite current collector thickness/density.

Response: PSI has provided additional details and detailed the projected impact on cell volumetric energy density.

Reviewer Comment: More continuous/pulse rate performance data and extended cycling information is needed to assess the impact of the higher loading cathodes on the cell performance.

Response: PSI demonstrated the HA coating enables improved continuous 5C rate performance, equivalent 1C and 2C performance, and stable full DOD cycling for thousands of cycles.

Partners/Collaborators

- CAMP Facility at Argonne National Laboratory:
  - Role: Characterization and Evaluation
  - Description: Scientists at ANL will assist in characterizing the physical structure and electrochemical performance of electrodes formed with multiple active materials using the PSI coating and electrode formulation techniques.

- SKC Powertech, Inc.:
  - Role: Cell builder/scale-up partner
  - Description: SKCP produced and delivered 3+Ah with the PSI coated cathode materials and electrode formulation techniques using their standard cell designs and construction procedures. These production efforts highlighted the reduced amount of solvent required when using the PSI coated materials. Additional cells will be built in the coming quarter.

Future Work: Upcoming Project Work and Challenges

Key Challenges

- **Challenge:** Demonstrate the targeted rate performance in Ah sized cell using the novel composite as the anode current collector.
  - PSI has demonstrated the ability to construct multi-layer pouch cells with the current collector addressing the electrochemical and mechanical stability of the connection/current collector.
- **Challenge:** Demonstrate the targeted cycle life performance in 3+Ah sized cells on operation to 4.4V or higher.
  - A second optimized version of 4.4V HA pouch cells is being built with adjustments to the electrolyte.

Future Work

- Construct Ah sized HA cell with the novel composite current collector.
- Construction of additional 3+Ah pouch cells.
  - Fully characterize the cycle life and rate performance of the pouch cells to demonstrate targeted 25% increase in energy density and required cycle life.

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

Summary

- Demonstrated HA coating enables production of electrodes with 98.5+% active material that deliver equivalent performance as standard commercial electrodes.
  - Increased electrode density enables >3-5% increase in cell energy density.
  - Demonstrated 50% reduction in the NMP volume required during electrode production.
  - Stable long term cycling achieved with 1700+ full DOD cycles and >90% capacity retention.
  - Successful scale-up and production of >20kg of coated material at coating cost <$0.2/kg.
  - Demonstrated applicability of HA coating to range of cathode materials including: $LiCoO_2$, NCA, and multiple NCM chemistries.
- Successfully demonstrated that stable high voltage operation increases cell energy by 10-14% when using HA coated material.
  - 400+ full DOD cycles achieved with 90% capacity retention.
- Successfully produced and demonstrated a composite anode current collector with <30% the mass of conventional copper foil.
  - Constructed multi-layer HA/composite current collector pouch cells and demonstrated efficient extended cycling.

25% increase in gravimetric energy density can be achieved through optimized cell design with the PSI technologies.