Solvent-free and Non-sintered 500 Wh/kg All Solid-State Battery

PI: Mike Wixom
(Presenter: Binsong Li)

Navitas Advanced Solutions Group

Project ID: BAT518

June 21-25, 2021

2021 DOE Vehicle Technologies Office Annual Merit Review

Overview

Timeline

- Project start: Oct. 2017
- Project end: Sep. 2021
- Percent complete: ~75%

Budget

- Total project funding: **$1.34M**
  - DOE Share: $1,025k
  - Contractor Share: $312k
- Funding for FY2020: $244k

Partners

- University of Maryland
- Oak Ridge National Lab
- **Project lead: Navitas**

Barriers

- Lack of large scale and low-cost solid-state battery (SSB) processing
- Low ionic conductivity and air stability of solid-state electrolyte (SSE) limit SSB application
- Low SSB performance due to unstable interphases of cathode/SSE and SSE/Li

Relevance

- **Impact:**

  Demonstrate a fabrication method for all solid-state Li metal battery to support Battery 500 program with similar performance to liquid-based systems but has:

  - Higher cell energy density due to high-quality thick cathode enabled by dry process;
  - Much lower cost through solvent-free electrode and non-sintered, stabilized sulfide solid-state electrolyte;
  - Improved safety due to usage of optimized solid-state electrolyte and strategies for stabilized interphases.

- **Objectives:**

  - The program aims to demonstrate a solvent-free and roll-to-roll fabrication process to enable large format solid state batteries incorporating stabilized sulfide solid-state electrolytes (SSE), high capacity cathodes and protected lithium metal anodes, that can deliver 500 Wh/kg specific energy and achieve life of 1000 charge/discharge cycles.

Milestones

Phase II, July 2020 – September 2021:

Month/Year	Milestone or Go/No-Go Decision Point	Status
July 2020	**1-2.** Solid-state electrolyte: Air stability 8 hours, Li ion conductivity 1.0 mS/cm **(Go/No-Go)**	**Complete**
September 2020	**2-1.** Cathode process down-selection	**Complete**
December 2020	**2-2.** 500g cathode powder production	**Complete**
December 2020	**3.** Thin Li anode, Li protection method verification	Delayed
December 2020	**4-2.** Large area and ultrathin SSE film	Delayed
July 2020	**5-1.** Preliminary cells assembled **(Go/No-Go)**	**Complete**
March 2021	**5-2.** 2.5 Ah pouch cell demonstrated	Delayed
June 2021	**5-3.** 12 Final test cells delivery	Delayed
March 2021	**6-1.** 500 Wh/kg cell demonstrated	Delayed
June 2021	**6-2.** Cycle life ≥ 1000	Delayed

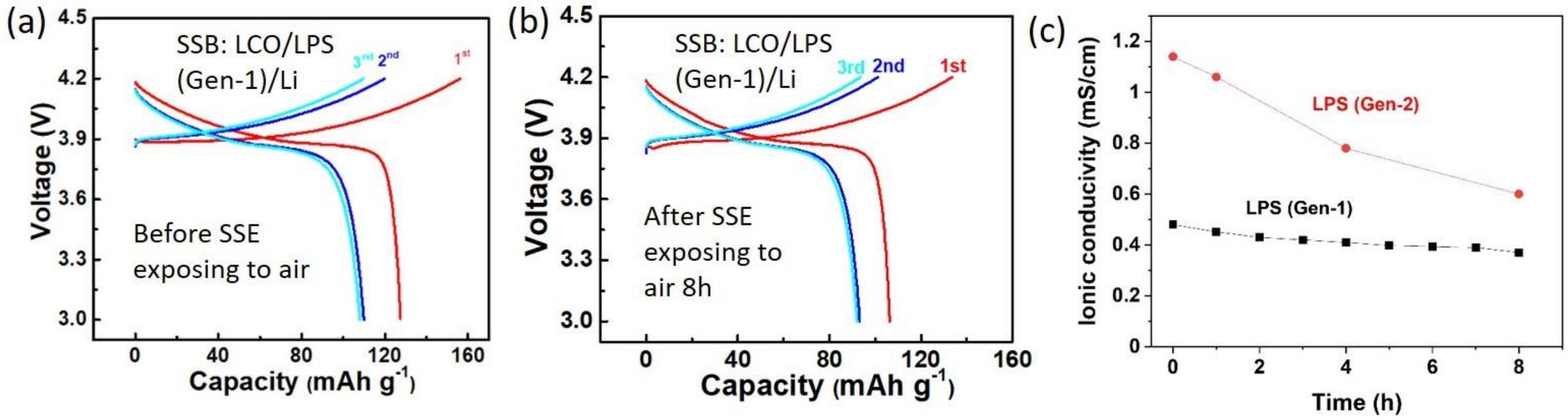
Approach

- **<u>Address Material Limitations:</u>**
  - Increase SSE ionic conductivity by new binder and additives, reduced binder content and SSE film thickness
  - Increase SSE air stability by additives and post-treatment
  - Increase cell energy by high loading cathode (now 5.6 mAh/cm^2) and thin SSE (60 µm so far)
  - Surface protection on both cathode (using 1, trying 2 more) and Li anode (wet chemistry and PVD) for longer cell cycle life

- **<u>Address Process Challenges:</u>**
  - Optimize dry process for thick cathodes and thin SSE to increase cell specific energy
  - Optimize dry process for large scale battery fabrication and low cost (2.5Ah Pouch Cells)
  - Non-sintered SSE to simplify battery fabrication and lower the cost

Accomplishments:

--- Solid-state Electrolyte Improvement



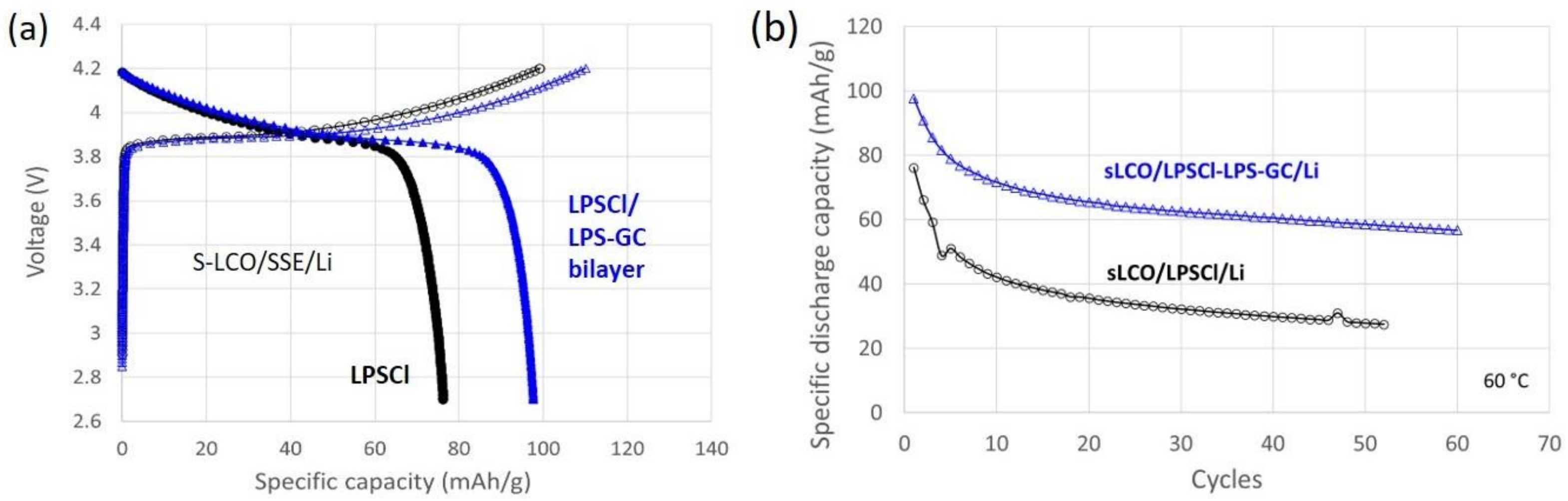
- The ionic conductivity of the Gen-1 LPS solid electrolyte was 0.5 mS/cm at 25 °C, which was stable for 8h exposure to dry air. The LCO SSB using LPS that has been exposed to dry air for 8 hours showed reasonable ~17% capacity decrease from 128 mAh/g to 106 mAh/g.

- The ionic conductivity of the LPS solid electrolyte increased from 0.5 mS/cm (Gen-1) to 1.1 mS/cm (Gen-2) by using a new dopant, and the conductivity was higher than G1 even after 8-hour exposure to dry air.

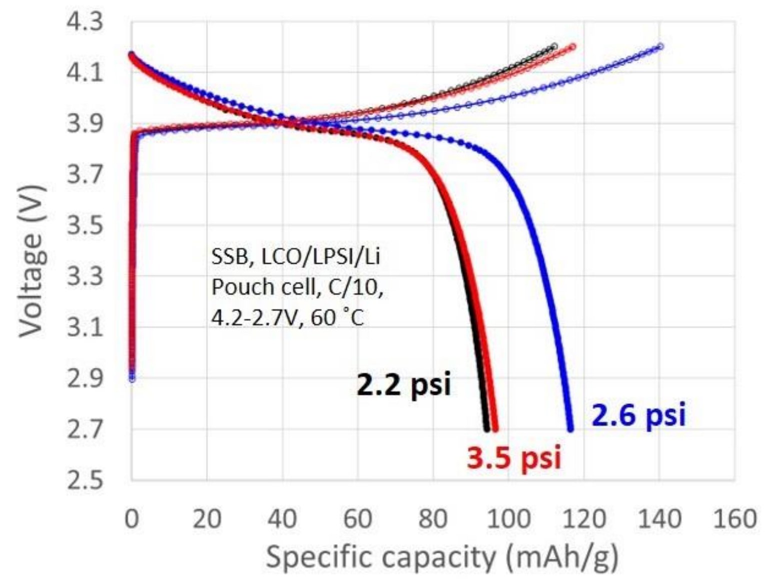
- The LPS synthesis was successfully scaled up to 200g/batch recently from 120g/batch, ensuring the processing of large amount of cathode and SSE composite for pouch SSB.

Accomplishments:

--- Solid-state Electrolyte Film and SSB Development



(Disk-in-pouch SSB: 0.1C rate, 2.7-4.2V, 60 °C)

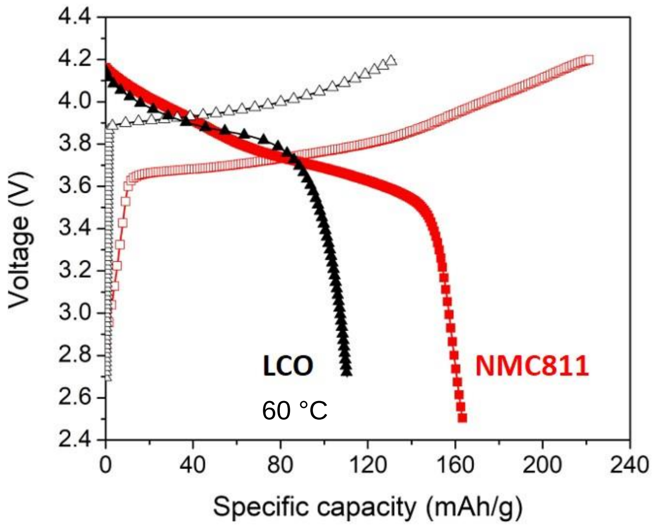


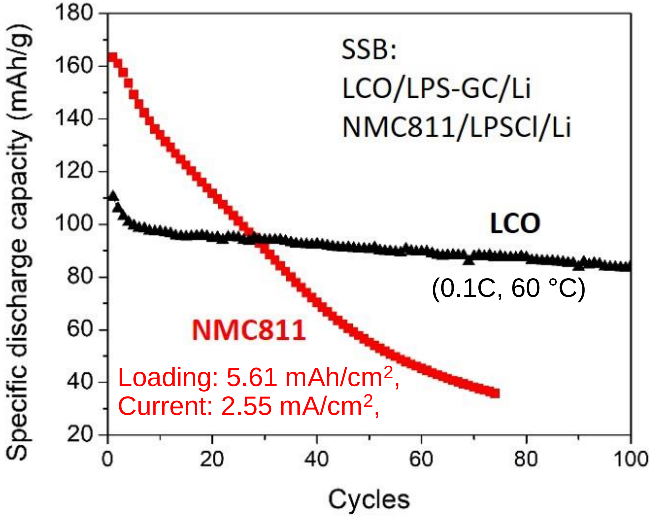
- A double layer SSE film with more stable LPS-GC SSE layer facing the lithium metal was adopted to mitigate side reactions and improve the SSB capacity and cycle life..

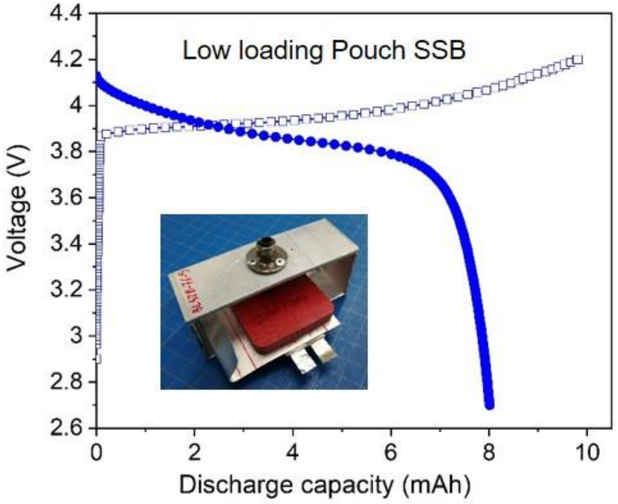
- The best external pressure on SSB has been tested and confirmed for the highest cell capacity.

Accomplishments:

--- Solid-state Battery Development







- Low loading (1 or 2 mAh/cm^2) up to 8 mAh Pouch SSB with LCO cathode has been delivered to DOE for testing.

- LCO as a stable cathode was used in most the development for screening purpose. A pouch SSB with small disk size cathode showed 1st specific discharge capacity of 110 mAh/g and a cycle life of 75 cycles at 0.1C, 2.7-4.2V.

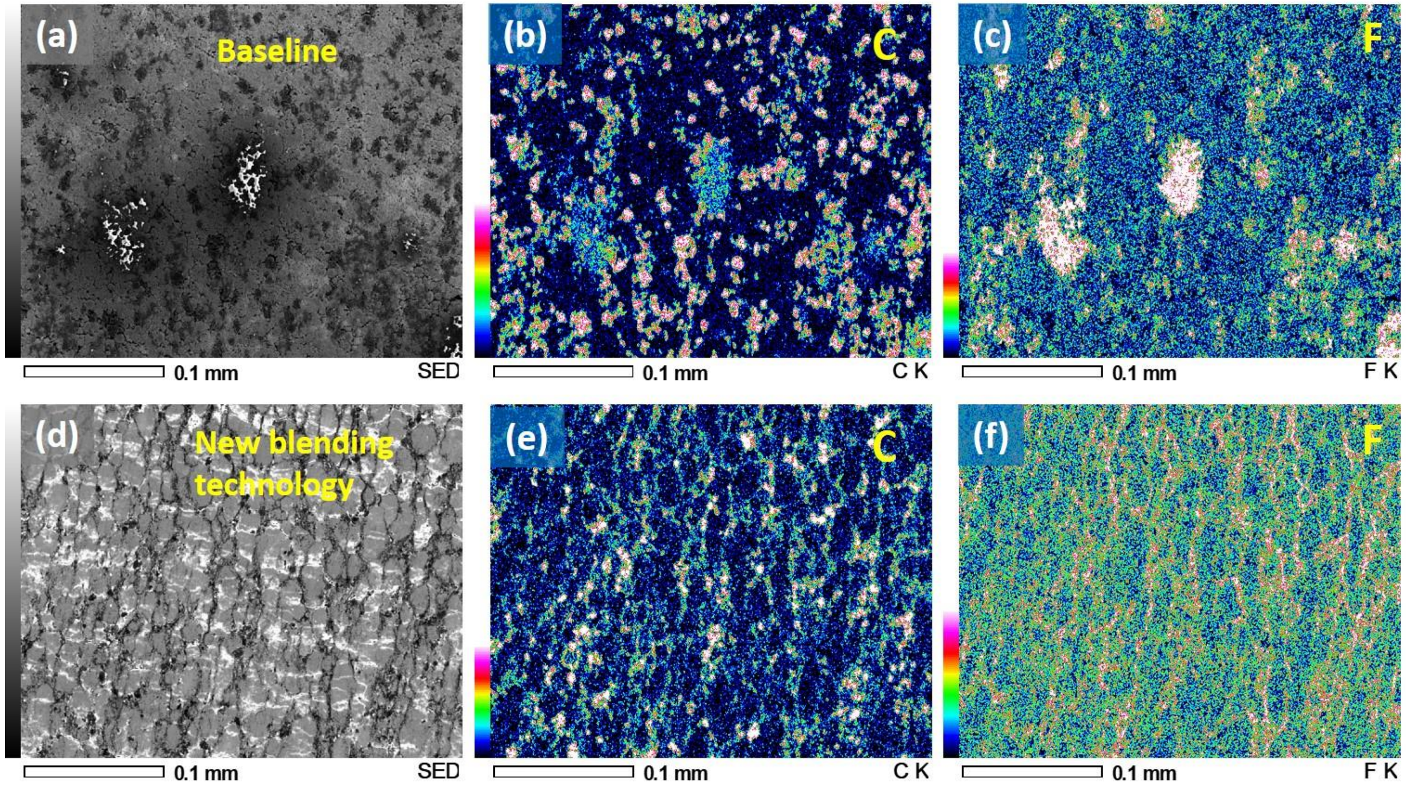
- SSB with high loading (5.6mAh/cm^2) NMC811 cathode showed higher specific discharge capacity of >160 mAh/g, but the capacity decayed fast due to unstable NMC811, high loading and fast rate ~0.5C.

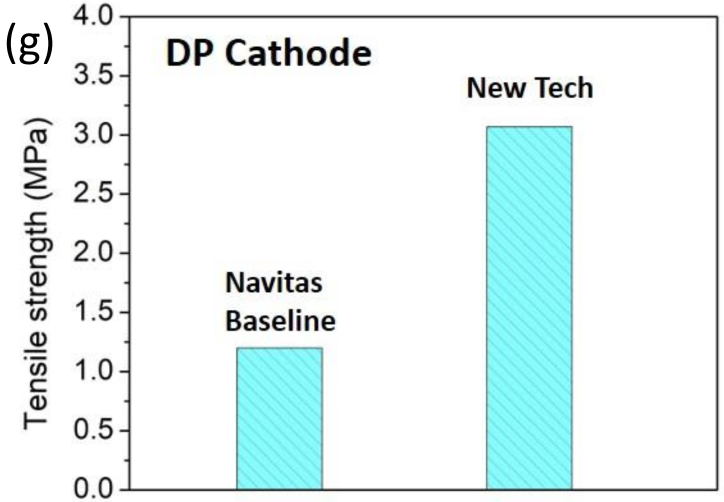
- Surface protection on both NMC811 and lithium anode is currently investigated with 3 options for each, which should improve the cell cycle life.

- New SSE with higher stability or double layer structure is also under testing to improve the cell cycle life.

Accomplishments:

--- New Advanced Dry Electrode Process





(NMC/Conductive carbon/Carbon additive/PTFE)

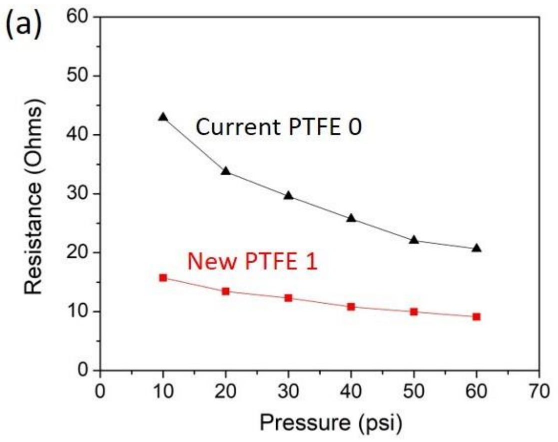
- **A new advanced dry electrode process** has been developed by Navitas and is being modified to use in SSB cathode and SSE film fabrication, which showed:
  - Enhanced carbon and PTFE binder dispersion (Figure e, f) comparing to current dry process (Figure b, c) to lower binder content for higher film ionic conductivity and much higher tensile strength (Figure g).
  - High-throughput for industrial application with low cost.
- **Formal patent application on Mar. 2021** initialed by Battery500 project and new equipment will be available in 2021.

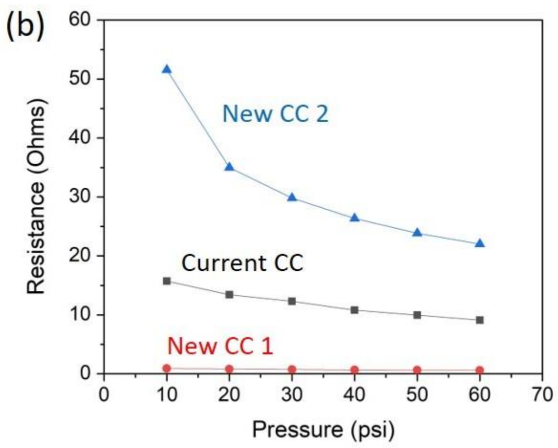
Accomplishments:

--- New Materials and Formulation for Dry Process

Cathode formulation:

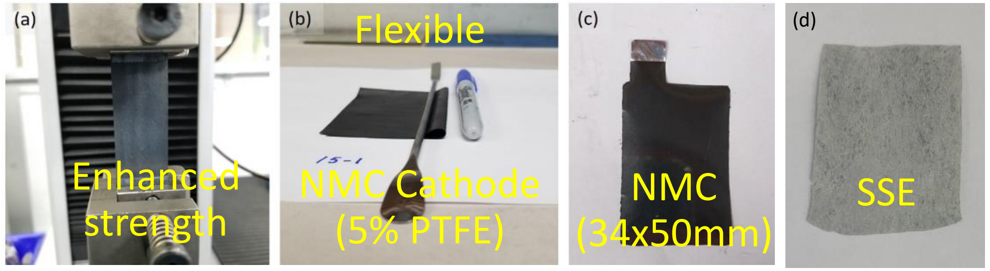
NMC/Conductive carbon (CC)/Carbon additive/PTFE = 95 : 1 : 2 : 5 wt%





Sample	Force @ Peak (N)	Young's Modulus (N/mm^2)	Tensile Strength (N/mm^2)
(1) PTFE 0, CC 0, std	1.64	306.04	0.71
(2) PTFE 1, CC 1, std	4.17	714.38	1.65
(3) PTFE 1, CC 2, std	2.92	244.60	1.20
(4) PTFE 1, CC 2, Adv	7.25	1455.54	**3.07**

- **New PTFE binder and conductive carbon (CC)** were screened. PTFE 1 and CC 1 were selected due to low film resistance.
- **The film tensile strength increased 132% by using new PTFE 1 and CC1** in standard dry cathode processing (from 0.71 to 1.65 N/mm^2). While if the new advanced blending technology is used, the tensile strength furtherly increased to 3.07 N/mm^2, corresponding to more than 4 times increase.
- **Navitas is working on applying these new materials and new blending technology to SSB** cathode and SSE film fabrication, and it is promising to solve the major issue of dry processed SSB on large film fabrication.



Selected Responses to Previous Year Reviewer's Comments

- **Reviewer 1** pointed out the best results were shown for the LCO cathode, while the most relevant to achieving 500 Wh/kg goals—NMC811 films with high loading—were prepared but have not been characterized.

  -- We added cell performance of NMC811 SSB with high loading of 5.6 mAh/cm^2. The specific capacity was ~160 mAh/g, much higher than that of LCO SSB, but the capacity decay was much faster and cycle life was only 11 cycles. We are trying new NMC811 surface coating, new Li anode coating and new dry processing technology to improve the cycle life.

- **Reviewer 2** has concern to reach to 1,000 cycles with 500 Wh/kg at room temperature due to no cycling performance on high loading NMC811 cathode cell and Li metal anode shows fast degradation at room temperature.

  -- The initial performance of high loading NMC811 SSB was much lower than the target. We see the goal is challenging, but we also see some new technologies exhibited promising results that we could use to mitigate the risk, such as 2 more new NMC811 surface coating, new Li anode surface protection that our partner UMD recently developed, and Navitas' new dry processing technology (formal patent application 2021). We expect these new technologies can improve the cycle life significantly.

- **Reviewer 3** was curious on the performance on various cell format of SSB (i.e., Swagelok cell --> coin cell --> disk-in-pouch --> pouch) and the pathway of the development.

  -- The Swagelok cell is the traditional format for lab scale. For easy operation and low-cost we developed SSB in coin cell, which had same performance. Disk-in-pouch cell was developed due to lack of large quantity of SSE and large film at early stage, which was a good simulation to large pouch cells since they have same performance and requirement, e.g, external pressure. I would like to mention that we could make large cathode or SSE film, but it requires higher binder content to prevent the free-standing film to break, which makes the film resistance much higher.

Collaborations and Coordination

University of Maryland (Sub) (C. Wang, S. Liu, H. Wan)

- Optimization of solid-state electrolyte (conductivity, stability)
- Demonstrate new solid-state battery chemistry
- Li anode protection by wet chemistry (Li-F rich and Li alloy containing SEI)

Oak Ridge National Laboratory (Sub) (D. Wood, J. Li, Z. Du)

- High purity thin Li anode preparation
- LiPON protected Li anode by PVD

Navitas Advanced Solutions Group (Prime) (M. Wixom, B. Li, M. Soueid, Kahla Sardo, B. Skelly, J. Hopkins)

Remaining Challenges and Barriers

- 500 Wh/kg target requires ideal condition of all components, such as high capacity cathode, thin SSE and stable interphase, which is very challenging. It may require to use a new equipment for the new blending technology we recently developed.
- Long cycle life is difficult to achieve for SSB with thick cathode.
- Scalable fabrication of ultrathin sulfide SSE film technically is very difficult. A stable SSE, a proper binder content and dispersion are required to retain a balance of high ionic conductivity and film mechanical strength.

Proposed Future Research

- **FY21:**
  - Optimize cathode and SSE formulation for balance of long cycle life and film strength
  - Scalable dry process of cathodes/SSE/Li stack towards 500Wh/kg pouch SSB
  - Improve interphase stability of cathode/SSE, Li/SSE for long cycle life
  - Assemble, test and optimize 2.5Ah pouch SSB

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

Summary

➢ **<u>Accomplishments:</u>**

- Optimized solid-state electrolyte with higher ionic conductivity (1.1 mS/cm) and dry air stability (8hr) that suitable for industry dry room process.
- Optimized dry electrode process with enhanced binder dispersion and reduced binder content.
- Fabrication and testing of SSB with high-loading NMC811 cathode (5.6 mAh/cm^2).

➢ **<u>Technical highlights:</u>**

- A new dry electrode process with enhanced binder dispersion and lower cost has been confirmed to bring much higher film tensile strength (up to 3 times), ensuring continuous R2R process. (Formal US Patent application, 2021)

➢ **<u>Impact toward VTO Objectives:</u>**

- This project will demonstrate a high energy density solid-state battery with feasibility to scale up to support Battery 500 program.
- The advanced dry electrode process can significantly reduce the cost of both solid-state batteries and liquid-electrolyte batteries.