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Name of Recipient:	Solid Power, Inc. 486 S. Pierce Ave., Suite E Louisville, CO 80027
Prime Recipient Type:	Private Company
Project Title:	All Solid State Batteries Enabled by Multifunctional Electrolyte Materials
Principal Investigator:	Pu Zhang
Team Members:	University of California, San Diego (Y. Shirley Meng)
Prime Recipient DUNS Number:	078313639
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Signature of Submitting Official:	

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Abstract

Solid Power has teamed with University of California San Diego to develop a high energy, long life, low cost, and safe all-solid-state-battery (ASSB). The battery is enabled by a multifunctional solid state electrolyte (SSE). The project enables scalable production of large format solid state batteries required by the vehicle market and building domestic battery manufacturers as leaders in the global vehicle ASSB production.

In the project, the multifunctional SSE materials have been developed and optimized with ionic conductivity ≥ 5 mS/cm and electrochemical stability 0 – 4.5V. SSE separator films have been coated by using a roll-to-roll process with thickness ≤ 40 μm. All-solid-state NMC-Li pouch cells containing the developed SSE have been assembled. A cycle life of > 750 at 100% DOD and 45 °C has been demonstrated in a full pouch cell.

Background

Objectives The project objective is to develop Li-metal solid batteries enabled by multifunctional SSEs for EV application. The ultimate goal is a scalable production of large-format ASSBs able to deliver ≥ 350 Wh/kg specific energy, ≥ 1000 cycle life, and ≤ $100/kWh cost.

Impact The project is enabling scalable production of large format all-solid batteries required by the vehicle market and building domestic battery manufacturers as leaders in the global vehicle ASSB production. The proposed technology addresses key limitations of state-of-the-art lithium batteries to meet DOE EV battery targets and accelerate their adoption as large-format EV batteries for sustainable transportation technology.

Approach A high-performance Li-metal all-solid-state cell is enabled by a multifunctional SSE. The new SSE: (1) has high conductivity (up to 10 mS/cm), (2) is stable against lithium metal and high-voltage cathode (0-4.5 V), (3) promotes uniform lithium plating (enabling > 2C charge rate), and (4) is compatible with large-scale manufacturing processes. The specific cell chemistry is the SSE with Li-metal anode and high-nickel-content Li-metal oxide cathode. The all-solid-state cell is assembled by scalable roll-to-roll (R2R) processes developed by Solid Power.

Collaborations The proposed team consists of Solid Power and a subcontractor, University of California, San Diego (UCSD). Solid Power (PI: Pu Zhang) has developed the multifunctional SSE and other cell components, assemble cells, and conduct cell tests. UCSD (Co-PI: Shirley Meng) has carried out material characterization by using advanced techniques such as XPS, cryogenic – scanning transmission electron microscopy (cryo-STEM) imaging, cryo-STEM energy dispersive X-ray spectroscopy (EDS), electron energy loss spectroscopy (EELS), and cryogenic – focused ion beam (cryo-FIB) milling.

Milestones

Period	Milestone	Type	Status	Completion Date
Year 1	**M1:** NMC cathode composition selected	Technical	100% Completion	12/31/2019
	M2: Cathode loading ≥ 3.5 mAh/cm^2	Technical	100%	03/31/2020
	M3: Critical current density ≥ 6 mA/cm^2	Technical	100%	06/30/2020
	• SSE ionic conductivity ≥ 3 mS/cm • Full cell cycle life ≥ 200	Go / No-Go	100%	09/30/2020
Year 2	**M1**: SSE stability 0 – 4.5V	Technical	100%	09/30/2021
	M2: Critical current density ≥ 18 mA/cm^2	Technical	100%	06/30/2021
	M3: R2R SSE film demonstration	Technical	100%	06/30/2021
	M4: 200 mAh pouch cell assembly	Technical	100%	06/30/2021
	• SSE conductivity ≥ 5 x 10^{-3} S/cm • Full cell cycle life ≥ 500	Go / No-Go	100%	09/30/2021
Year 3	**M1:** Charge rate ≥ 2C	Technical	100%	12/31/2021
	M2: SSE film ≥ 10 m and ≤ 40 μm	Technical	100%	03/31/2022
	M3: Pouch cell ≥ 2 Ah	Technical	100%	06/30/2022
	M4: Cycle life ≥ 1000	Technical	75%	09/30/2022

Accomplishments:

1. Multifunctional solid state electrolyte (SSE) development

Solid Power's halogenated LPS material was used as a starting point. Li_2S, P_2S_5, a halogen and other selected dopants were ball-milled to form glassy sulfide electrolytes by using a planetary ball mill. A subsequent heat-treatment was conducted to obtain glass-ceramic solid electrolytes.

The composition and process of the electrolyte materials were investigated extensively by using "Design of Experiments" (DOE) technique. An L18 orthogonal matrix was applied, which consisted of 18 experiments and 8 control factors. The control factors included dopant parameters, milling parameters, and heat treatment parameters as shown in Table 1. The powder pre-treatment factor had 2 levels and all other 7 factors had 3 levels. Li ion conductivity, critical current density (CCD) with Li, and long term cyclability of the electrolyte were used as the key criteria to qualify the electrolyte materials.

The final optimized electrolyte material has shown a Li ionic conductivity of 6 mS/cm at 25°C and 2 mS/cm at 0°C. A CCD of 20 mA/cm^2 at 70°C was also demonstrated with the electrolyte. Figure 1 shows the electrolyte Li ion conductivity and CCD.

Table 1. Process parameter design control factors
(■ indicates the baseline conditions developed previously)

Control Factors		Level 1	Level 2	Level 3
1	Dopant type	A	B	C
2	Doping level	d_1	d_2	d_3
3	Powder pre-treatment	Yes	No	
4	Milling speed	M_1	M_2	M_3
5	Milling time	t_1	t_2	t_3
6	Media to powder ratio	R_1	R_2	R_3
7	Process temperature	T_1	T_2	T_3
8	Process atmosphere	A_1	A_2	A_3

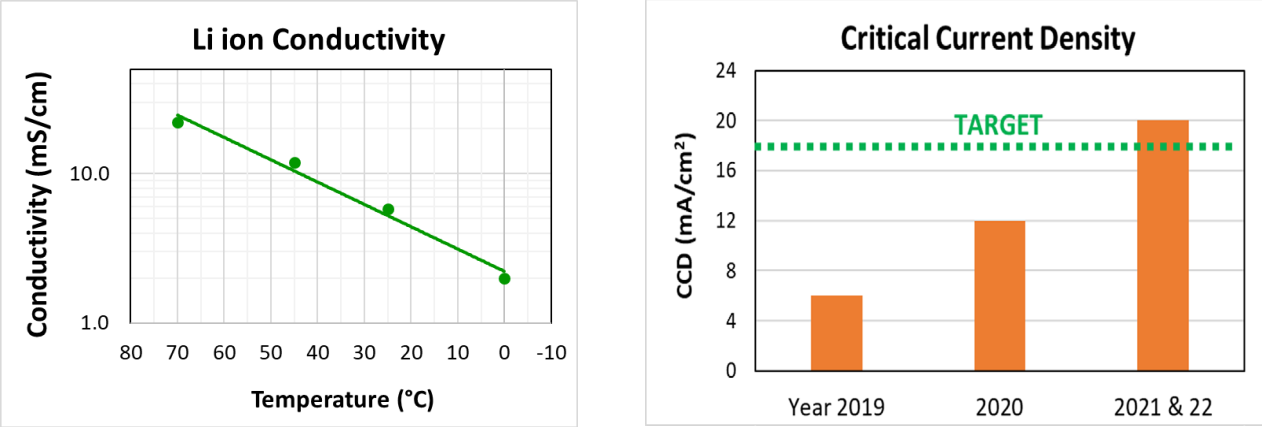

Figure 1. The electrolyte developent progress: Li ion conductivity at varous temperatures and CCD at 70°C

The high voltage stability of the new electrolyte was evaluated in a Li-NMC full cell. A conventional LPS electrolyte was also tested as a baseline. When charged to 4.5V, C/10 – C/10, and 45°C, the new electrolyte cell was stable while the baseline electrolyte cell showed 20% resistance growth after 10 cycles. The new electrolyte cell also showed 60% lower resistance than the baseline cell. Figure 2 shows the cell resistance growth comparison and confirms the new electrolyte's high voltage stability.

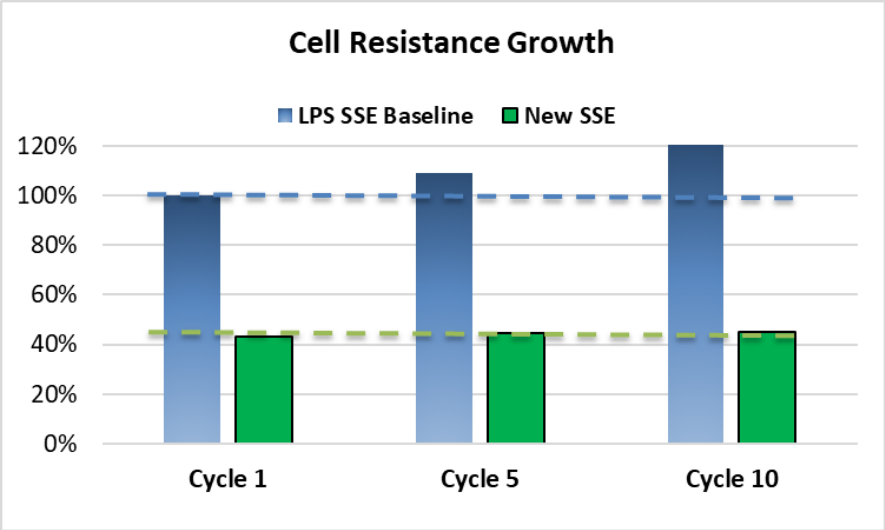

Figure 2. Li/SSE/NMC full cell resistance growth: new electrolyte vs. baseline electrolyte. The cells were tested at 2.5 – 4.5V, C/10 – C/10, and 45°C. The 4.5V stability of the new electrolyte is demonstrated.

2. SSE separator fabrication

The SSE separator coating process has been developed at pilot scale. A separator slurry was prepared by mixing the SSE powder, a binder, and a solvent by using a planetary high-shear mixer. The slurry was then cast on a carrier film on a slot-die coater. The separator length and thickness target (≥ 100 m and ≤ 40 µm) was achieved. Figure 3 shows a separator coating on the slot-die coater with a thickness of 30 µm measured by SEM.

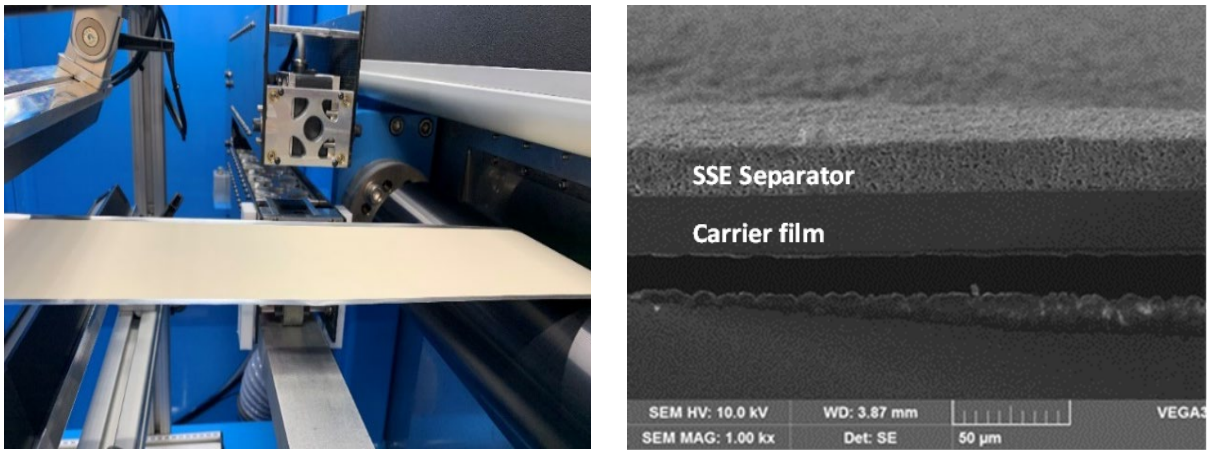

Figure 3. A solid-state-electrolyte separator coating by a roll-to-roll process

3. Full pouch cell demonstration

Pouch cell assembly

A thin Li foil (35 μm) anode is used as received. An NMC622 cathode is selected for the pouch cell. Both the cathode and the SSE separator are slurry-casted on a slot-die coater. A tri-layer of anode/SSE/cathode is then laminated. A single piece of the tri-layer film or multiple pieces are being used to form a single-layer or multilayer cell stack. After welding tab to the stack and sealing the tabbed stack into a pouch, a pouch cell is formed. The cell capacity ranges from 6 mAh (i.e., a single-layer-pouch cell) to 2 Ah (i.e., a large format multilayer cell) in the project.

Cycle life

A single layer pouch cell (at 6 mAh) was assembled for the performance demonstration. The cell contained an NMC 622 composite cathode (at 3 mAh/cm^2), a thin Li metal anode, and a SSE separator. When tested at C/5 - C/5, 2.8 - 4.2V, and 45°C, the cell showed 87% capacity retention after 750 cycles (in Figure 4). It should be noted that the cell contained a pure Li anode. Surface modification of the Li anode is expected to improve the cycling stability further in the future development.

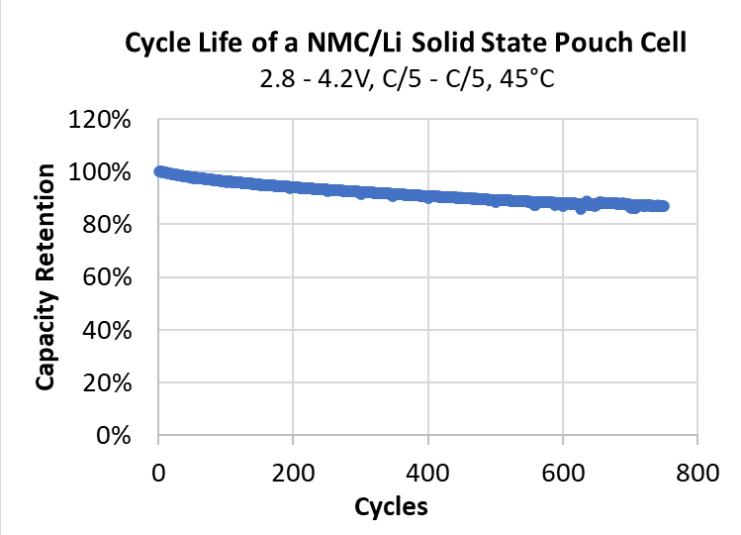

Figure 4. Cycle life of an NMC/Li metal solid state pouch cell

Calendar life

The calendar life was also evaluated in the 6 mAh Li-NMC pouch cell. The cell was formed at 2.5V - 4.2V at C/10 and 45°C. It was then charged to 4.2V at C/10 & 45°C (100% SOC) and stored at 60°C for 30 days. After the storage, the cell was discharged to 2.5V ("capacity retention"), recharged to 4.2V, and discharged again to 2.5V ("capacity recovery") at C/10 and 45°C. As shown in Figure 5, the cell capacity retention is 87% and capacity recovery is 95%, when compared to the initial discharge capacity after the storage at 60°C for 30 days. Both the capacity retention and recovery from the ASSB cell are comparable to or better than those with

the conventional Li ion battery cell at the same testing conditions. The storage result confirms the long term stability of the SSE and suggests the ASSB cell having a calendar life > 10 years.

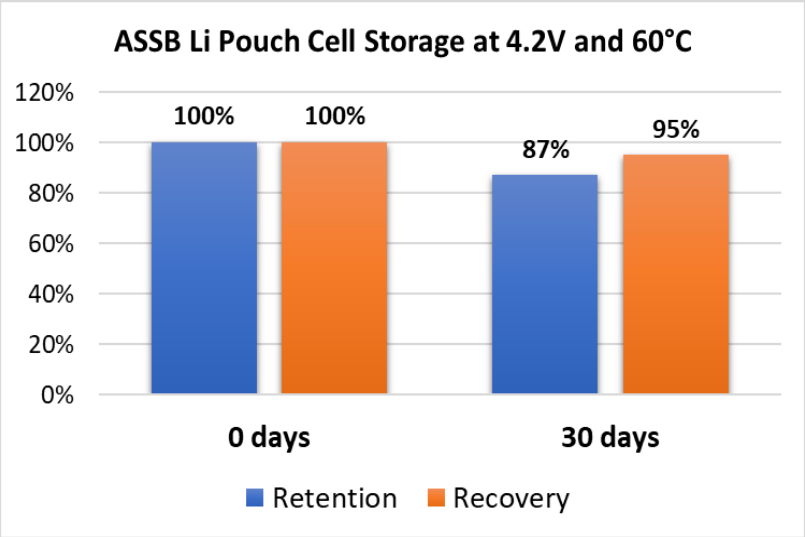

Figure 5. Storage test of a Li/SSE/NMC solid state pouch cell at 60°C

Rate capability

Rate capability of the pouch cell was also evaluated by charging and discharging symmetrically from C/10 to 2C at 70°C. As shown in Figure 6, the cell retained 78% capacity at 2C, when compared to C/10. This result indicates that a solid state Li metal cell is capable of fast charging at an elevated temperature. The future development focus will be demonstrating the fast charge capacity at lower temperatures.

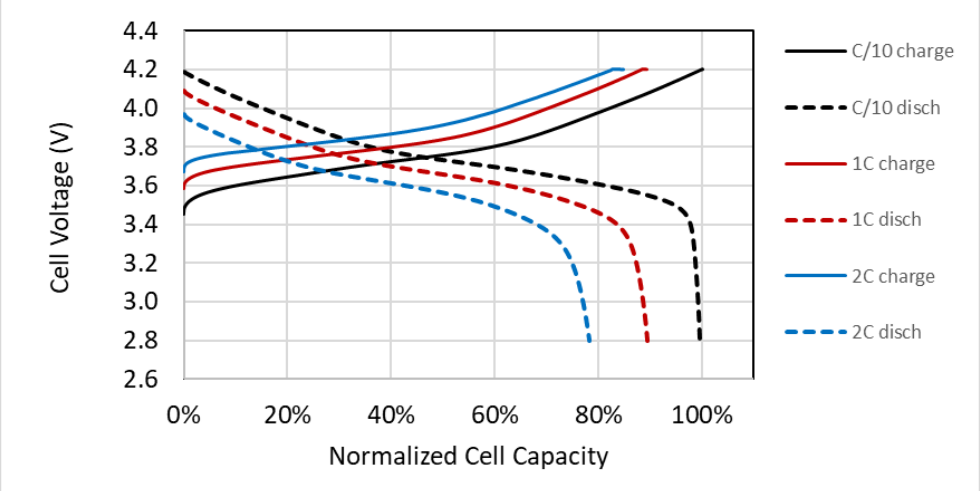

Figure 6. Rate capacity of a Li/SSE/NMC solid state pouch cell at 70°C

4. All-solid-state-battery (ASSB) cell design

ASSB cell platform

The solid state electrolyte (SSE) replaces all of the liquid electrolyte in a solid-state cell, but many cell designs are possible under the solid-state umbrella. Solid Power's SSE materials enable an entire new cell platform that can incorporate a wide variety of existing and future anode and cathode materials (Figure 7).

On the anode side, Li metal is an ultimate solution as it stores ultra-high energy per unit mass and it delivers low cost long-term. Silicon is also attractive as it offers the quickest path to meeting all of the charge rate and low-temperature cycling requirements for future electric vehicles in low-cost EV scale cells, while delivering similar volumetric energy density to Li metal if enough Si material can be packed into the anode layer.

On the cathode side, the sulfide electrolytes can be paired with the same lithium nickel-manganese-cobalt oxide (NMC) cathode materials being used today in EVs, which is important for near-term market introduction. Conversion reaction type cathode chemistries such as iron sulfide, enabled by the sulfide SSE materials, would provide further gains in specific energy while being inexpensive and sustainable.

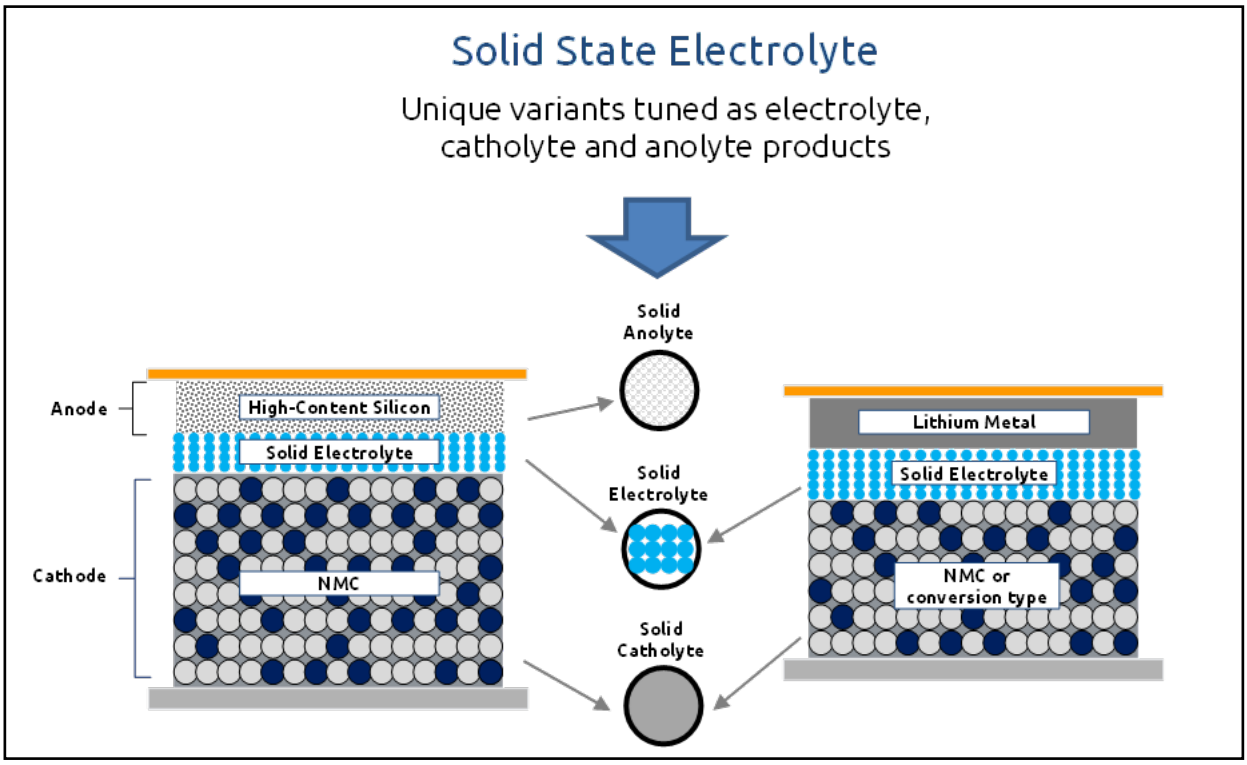

Figure 7. The sulfide SSE materials enabling a flexible platform that incorporates a variety of anode and cathode materials for a high energy and low cost all-solid-state-battery cell

ASSB cell energy projection

In this project we have demonstrated a multifunctional SSE separator at 40 µm, an NMC 622 cathode at 3.2 mAh/cm^2 loading, and a stand-alone Li metal anode at 35 µm in a full pouch cell.

By applying the current cell parameters to an EV-relevant pouch cell at 100 Ah, the cell specific energy and energy density are projected be 350 Wh/kg and 810 Wh/L, respectively.

In the future development beyond the project, we plan to validate an NMC 811 cathode in the ASSB cell. By applying the NMC 811 cathode, increasing the cathode loading to 3.5 mAh/cm^2, and reducing the separator thickness to 35 µm. An EV cell with energy densities at 400 Wh/kg and 950 Wh/L is achievable.

The key cell design parameters and projected EV cell specific energy & energy density in the current generation and next generation technologies are summarized in Table 2.

Table 2. ASSB Li pouch cell design parameters and projected energy densities

Timeline	Key Cell Design Parameter				100 Ah EV Cell Specific Energy and Energy Density (Projected)	
	Cathode Type	Cathode Loading (mAh/cm^2)	Li Thickness (µm)	Separator Thickness (µm)	Specific Energy (Wh/kg)	Energy Density (Wh/L)
Current Project	NMC 622	3.2	35	40	**350**	**810**
Next Generation	NMC 811	3.5	35	35	**410**	**950**

Conclusion

Multifunctional solid-state-electrolyte (SSE) materials have been developed and optimized with high conductivity and electrochemical stability. SSE separator films have been coated by using a roll-to-roll process. All-solid-state NMC-Li pouch cells containing the developed SSE have been assembled. A cycle life of 750 at 100% DOD and 45 °C has been demonstrated in a full pouch cell. A calendar life > 10 years and a specific energy > 350 Wh/kg have been projected in an EV cell format.