

**Final Scientific/Technical Report**  
**Developing Materials for High-Energy-Density  
Solid State Li-S Batteries**

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## Executive Summary

Solid-state lithium-sulfur batteries are considered to be the next-generation power source for vehicle applications due to their high energy density (up to 3 times more than current Li-ion) and low cost of sulfur (100 times less than conventional cobalt oxide). However, issues with solid-state electrolyte stability, conduction pathways in the sulfur cathode, and the interface between solid-state electrolyte and sulfur cathode must be solved in order to allow commercialization. This project demonstrates the creation of new, advanced materials which overcome these inherent issues.

Current approaches based on polymer or liquid electrolyte with additives have been able to improve stability at the cost of efficiency. In contrast, by creating new hybrid materials it is possible to achieve both stability and efficiency. We developed a novel sulfide-based solid electrolyte, a sulfur-carbon composite cathode, and explored various additives to stabilize the interface of solid electrolyte and cathode. After the desired properties were achieved, these materials were integrated into a high-performance solid-state battery, bringing low-cost high-energy solid-state lithium-sulfur batteries one step closer to reality.

## Project Background

Lithium-sulfur (Li-S) batteries have received great attention due to the potential to achieve high energy density at low cost for electric vehicle applications. The significant challenge for conventional Li-S batteries using liquid electrolyte is its rapid capacity fading that arises from the polysulfide dissolution, the resultant shuttling effects, and deterioration of Li metal. Novel Li-S battery components have been developed to address the critical challenges spanning from the sulfur cathode, electrolyte and additives to anode and cathode protection for improvement of cycling stability.

Yet, those approaches succeed in improving cycling but not eliminating the polysulfide dissolution and shuttling effects. Recent developments in solid-state electrolyte have renewed the interest in solid-state Li-S battery to replace liquid electrolyte. In principle, given that Li-S reaction intermediates will not form liquid component in solid-state electrolyte, the solid-state Li-S battery can fundamentally remedy the polysulfide dissolution and shuttling effects to achieving better cycle performance.

While promising, the solid-state Li-S batteries still have challenges in achieving high energy density and excellent cycling stability while maintaining good rate performance. The major issues come from the following aspects:

- 1) Due to insulating nature of sulfur, mixing sulfur with carbon material is essential to facilitate electronic conduction in the cathode of solid-state Li-S batteries. Therefore, both selection of carbon materials and control of sulfur distribution in sulfur-carbon composites are important to increase specific capacity and improve rate performance in solid-state Li-S batteries. Development of new carbon-sulfur composite with controlled distribution of sulfur can increase specific capacity of sulfur and sulfur utilization at high mass loading based on our preliminary results.

- 2) Interfaces between sulfur-carbon composite and solid-state electrolyte are pivotal to achieving high specific capacity and long cycling stability in solid-state Li-S batteries. It is

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desirable to develop an innovative approach to control and tune interface within cathode so as to enable complete and highly reversible lithiation/delithiation of sulfur.

3) Glass and glass-ceramic sulfide-based solid-state electrolytes are compatible with sulfur cathodes and widely used in solid-state Li-S batteries. However, the conductivity of the glass and glass-ceramic solid-state electrolyte is as low as  $10^{-4}$  S/cm. In addition, control of morphology and interfacial properties of the solid-state electrolyte is lacking in construction of carbon-sulfur-electrolyte cathode for solid-state Li-S batteries.

Resolving these issues requires the development of new materials: a novel carbon-sulfur composite with controlled structure and distribution; a solid additive to tune the interfacial properties of solid-state electrolyte; and an advanced sulfide-based solid-state electrolyte with improved moisture stability and high Li-ion conductivity.

## Project Objectives & Accomplishments

The objective of this project is to develop materials, which involves advanced carbon materials, solid additives, and sulfide-based solid-state electrolytes, and acquire knowledge for lithium-sulfur all-solid-state batteries (Li-S ASSBs) in order to meet DOE's targets for high-energy-density battery ( $\geq 350$  Wh  $\text{kg}^{-1}$  at C/3) with long cycle life (C/3 deep discharge and  $< 20$  % energy decay) and low cost ( $\leq \$100$  kWh $^{-1}$ ). Li-S ASSBs with high areal sulfur loading ( $\geq 5$  mg  $\text{cm}^{-2}$ ) and high sulfur content ( $\geq 50$  wt% in cathode), pairing with lithium or lithium alloy anode, shall deliver a high initial specific capacity of over 1200 mAh  $\text{g}^{-1}$  at high charge/discharge rate ( $> 0.3$  C) for 500 cycles with over 80% capacity retention.

Specifically, approaches to realize the project objectives include the following:

- (1) development of new carbon material with unique structure, high surface area, and large pore volume;
- (2) development of new S-C and S-C-M<sub>x</sub>S<sub>y</sub> materials (M = Li, Co, Ti, Mo, etc.) to facilitate electron/ion transport;
- (3) development of novel additives to tune interfacial behavior among components in the cathode;
- (4) development and optimization of new SSE through cation and anion doping with superior properties such as high ionic conductivity, good moisture, and stability; and
- (5) diagnostics, characterization, and cell tests on the developed new material or advanced sulfur cathode.

Milestone	Type	Description	Actual Results
<b>Optimal cation and anion co-doped sulfide solid state electrolyte demonstration</b>	Technical	Demonstrate cation and anion-doped sulfide solid electrolyte with ionic conductivity of above 5 mS $\text{cm}^{-1}$ , improved stability against moisture at room temperature.	New sulfide solid electrolyte Li <sub>3</sub> PS <sub>4</sub> -2LiBH <sub>4</sub> with ionic conductivity of 6.0 mS/cm at room temperature.

<b>Sulfur-Carbon-M<sub>x</sub>S<sub>y</sub> hybrid material demonstration</b>	Technical	Demonstrate sulfur cathode (sulfur content = 50 wt%, 1.5 mgSulfur cm <sup>-2</sup> ) fabricated using the developed carbon-M <sub>x</sub> S <sub>y</sub> hybrid material with a stable capacity of above 1000 mAh g <sup>-1</sup> at 0.3 C at 60 °C for 50 cycles.	Sulfur cathode of 50:10:24 sulfur:carbon:solid electrolyte mass ratio (60 wt% S) with capacity 905.3 mAh/g at 5.16 mg/cm <sup>2</sup> and 0.1 C, 60 °C for 50 cycles.
<b>EAMs additives demonstration</b>	Technical	Demonstrate sulfur cathode (sulfur content = 50 wt%, 1.5 mg <sub>Sulfur</sub> cm <sup>-2</sup> ) fabricated using EAMs as additives (less than 3 wt%) with a stable capacity of above 1000 mAh g <sup>-1</sup> at 0.3 C for 50 cycles at 60 °C.	Additive TiS <sub>2</sub> at 0.38 wt% in sulfur cathode (39 wt% S, 1.5 mg/cm <sup>2</sup> ) demonstrated capacity 1200 mAh/g at 0.3 C for 50 cycles at 60 °C.
<b>Optimal solid-state Li-S cells demonstration</b>	Technical	Demonstrate all-solid-state Li-S batteries with large areal sulfur loading (≥ 5 mg cm <sup>-2</sup> ) using additives (less than 3 wt%), high sulfur content (≥ 50 wt% in cathode), high specific capacity (1200 mAh g <sup>-1</sup> at over 0.3 C discharge/charge rate) and superior cycle life (< 20% capacity decay in 500 cycles) at room temperature.	Test cells with mass loading 3.0 mg/cm <sup>2</sup> using developed materials achieved a capacity of 1068 mAh/g at 0.2 C, with excellent cycle life (14% capacity decay at 500 cycles) at 60 °C.

## Summary of Project Activities

### 1) Sulfide-based solid electrolyte development

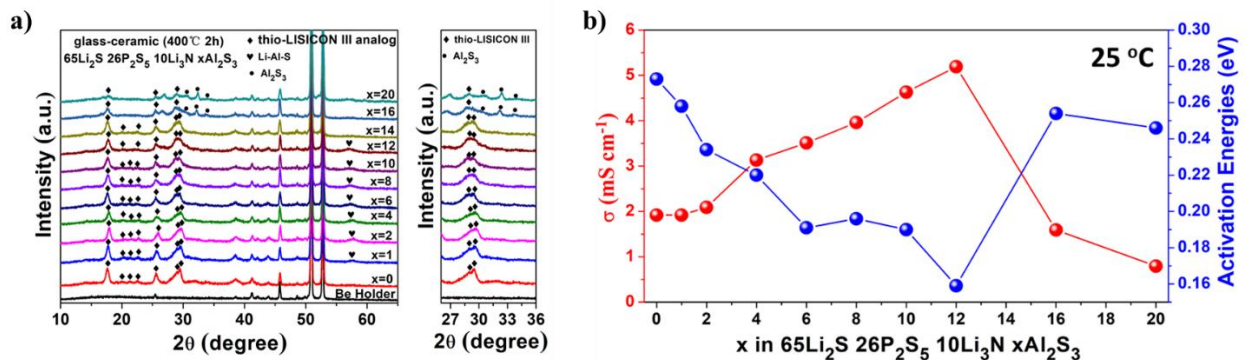
Solid electrolyte is one of the key components that shall affect sulfur cathode performance in Li-S ASSBs. It is crucial to gain knowledge of its impacts and desired properties as well as developing solid electrolytes with high ionic conductivity at room temperature.

To gain more knowledge of solid electrolytes and understand its impact on sulfur cathode performance, we prepared various glass and glass-ceramic solid electrolytes, which includes 75Li<sub>2</sub>S·25P<sub>2</sub>S<sub>5</sub> glass solid electrolyte (LPS), Li<sub>6</sub>PS<sub>5</sub>Cl glass solid electrolyte, and Li<sub>7</sub>P<sub>2</sub>S<sub>8</sub>Br<sub>0.5</sub>I<sub>0.5</sub> glass-ceramic solid electrolyte using both dry ball milling approach and wet ball milling approach. We found that the wet ball milling approach which utilizes heptane as dispersing agent is more favorable for the formation of smaller solid electrolyte particles compared with dry ball milling approach) and therefore more favorable for uniform mixing with sulfur-carbon composite and

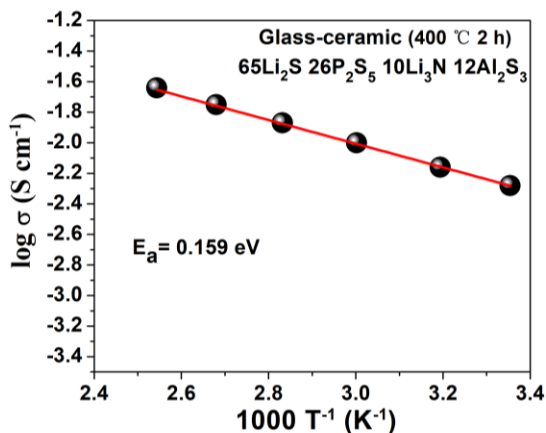
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cathode performance. We also found that for LPS glass solid electrolyte prepared from wet ball milling process showed higher ionic conductivity than electrolyte prepared from dry ball milling approach ( $3.8 \times 10^{-4} \text{ S cm}^{-1}$  at  $25^\circ\text{C}$  for wet process,  $3.2 \times 10^{-4} \text{ S cm}^{-1}$  at  $25^\circ\text{C}$  for dry approach).

Besides gaining more knowledge of solid electrolytes that have already been developed in literature, we also developed a new solid electrolyte,  $65\text{Li}_2\text{S}-65\text{Li}_2\text{S}-26\text{P}_2\text{S}_5-10\text{Li}_3\text{N}-12\text{Al}_2\text{S}_3$  (LPSNAI-12) glass-ceramic solid electrolyte, by incorporating nitrogen and aluminum in the electrolyte. The electrolyte demonstrated high ionic conductivity at  $25^\circ\text{C}$  ( $5.19 \text{ mS cm}^{-1}$  at  $25^\circ\text{C}$ ), low activation energy of  $0.159 \text{ eV}$ , and superior stability against lithium metal anode. To synthesize this material, we optimized the content of  $\text{Al}_2\text{S}_3$  in the electrolyte. The detailed X-ray diffraction (XRD) spectra, ionic conductivity at  $25^\circ\text{C}$  and activation energy of solid electrolytes with different  $\text{Al}_2\text{S}_3$  content are summarized in Figure 1. It was observed that for  $65\text{Li}_2\text{S}-65\text{Li}_2\text{S}-26\text{P}_2\text{S}_5-10\text{Li}_3\text{N}-12\text{Al}_2\text{S}_3$  glass-ceramic solid electrolyte the thio-LISICON III analog phase is precipitated on XRD spectra. The detailed ionic conductivity at different temperature is summarized in Figure 2. The origin of such high ionic conductivity might be related to the incorporation of nitrogen and aluminum in the structure. Formation of P-N, P=N and Li-Al-S was observed by x-ray photoelectron spectroscopy (XPS). In addition to the ionic conductivity and structure evaluation of the synthesized solid electrolyte, its stability against lithium metal has also been studied. According to the Li/LPSNAI-12/Li symmetric cell test, the developed new solid electrolyte is stable against lithium metal. Furthermore, the polarization resistance at  $0.6 \text{ mA cm}^{-2}$  is just  $25 \Omega \text{ cm}^{-2}$ . And long-term stable cycling of the symmetric cell at  $0.6 \text{ mA cm}^{-2}$  and  $0.6 \text{ mAh cm}^{-2}$  for over 1000 hours have been achieved. All of these superior properties demonstrate that the solid electrolyte we developed,  $65\text{Li}_2\text{S}-65\text{Li}_2\text{S}-26\text{P}_2\text{S}_5-10\text{Li}_3\text{N}-12\text{Al}_2\text{S}_3$ , is a good candidate for future Li-S ASSBs.

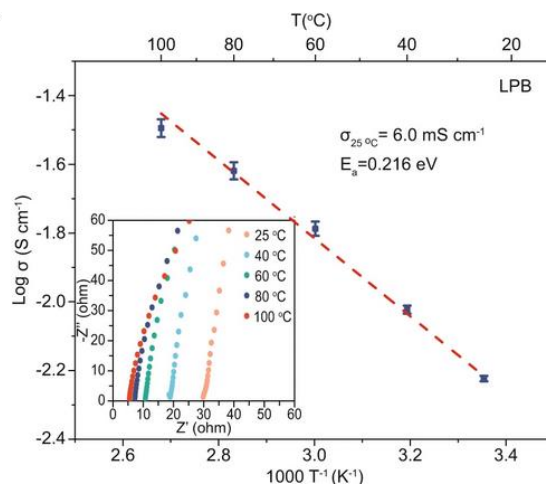


**Figure 1.** a) X-ray diffraction (XRD) spectra and b) ionic conductivity at  $25^\circ\text{C}$  and activation energy of various  $65\text{Li}_2\text{S}-65\text{Li}_2\text{S}-26\text{P}_2\text{S}_5-10\text{Li}_3\text{N}-x\text{Al}_2\text{S}_3$  (LPSNAI-x) glass-ceramic solid electrolyte ( $x=0 - 20$ ).



**Figure 2.** The Arrhenius plot for the measured ionic conductivity of LPSNAI-12 glass-ceramic solid electrolyte.

Furthermore, we continued to develop a new sulfide solid electrolyte  $Li_3PS_4-2LiBH_4$  (LPB), synthesized via a liquid-phase method with a low measured density ( $1.491 g cm^{-3}$ ), and high ionic conductivity ( $6.0 mS cm^{-1}$  at  $25 ^\circ C$ ). Its extremely low density serves to enhance the gravimetric energy density when considered as part of a cell. Measurement of a cold-pressed LPB pellet showed an ionic conductivity of  $3.8 mS/cm$  at  $25 ^\circ C$ . Hot pressing of the pellet was found to increase the density up to 91.5%. Figure 3 shows the measurement of ionic conductivity for a hot-pressed LPB pellet, namely  $6.0 mS/cm$  at  $25 ^\circ C$ , with an activation energy of  $0.216 eV$ . The solid electrolyte was created via liquid-phase synthesis, combining  $Li_2S$ ,  $P_2S_5$ , and  $LiBH_4$  in tetrahydrofuran solvent. This leads to a low synthesis temperature of  $160 ^\circ C$ , lower than solid electrolytes with comparable ionic conductivity. The structure of LPB was investigated using X-ray powder diffraction. The cubic argyrodite phase crystalline  $Li_{6-x}PS_5-x(BH_4)_{1+x}$  ( $-1 < x \leq 1$ ) was identified, corresponding to the high ionic conductivity. However, the lower density compared to the theoretical is due to the amorphous phase.



**Figure 3.** Arrhenius plot for cold-pressed LPB SE pellet and the corresponding Nyquist impedance plot of the pellet tested at different temperatures (i.e., 25, 40, 60, 80, 100 °C). The error bar represents the standard deviation of the measured ionic conductivity.

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Solid-state nuclear magnetic resonance (NMR) spectroscopic characterization of LPB was further carried out to attain more details of the LPB's local chemical structure and discern the impurities. As shown in the  $^{31}\text{P}$  NMR spectra (Figure 4), three distinct peaks, I, II, and III, were observed. Peak I can be deconvoluted into two lines, implying the diverse chemical environment of  $\text{PS}_4^{3-}$  tetrahedrons in the argyrodite  $\text{Li}_{6-x}\text{PS}_{5-x}(\text{BH}_4)_{1+x}$  induced by the disorder in  $\text{S}^{2-}/\text{BH}_4^+$  sublattices. This disorder is favorable for improving the ionic conductivity in argyrodite SE, which might explain the good  $\text{Li}^+$  conductivity in LPB.

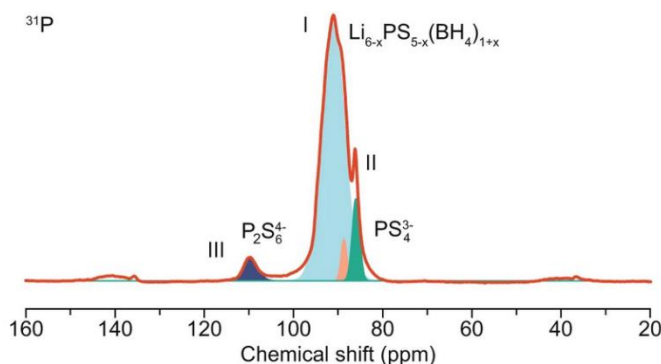


Figure 4.  $^{31}\text{P}$  MAS NMR of LPB powder.

The Li/LPB/Li symmetric cell showed stable cycling up to a current density of  $1.0 \text{ mA/cm}^2$ , demonstrating stability against Li metal. At  $1.0 \text{ mA cm}^{-2}$ , the polarization resistance of the LBP is  $38 \Omega \text{ cm}^{-2}$ . All these properties make LBP an ideal solid electrolyte for further Li-S development.

## 2) Carbon materials for sulfur cathode

Within the sulfur cathode, the carbon additive is the key component for constructing an electronic pathway. Since sulfur and solid electrolytes are insulating, the effectiveness of the electronic conductive network is determined by the carbon additive utilized.

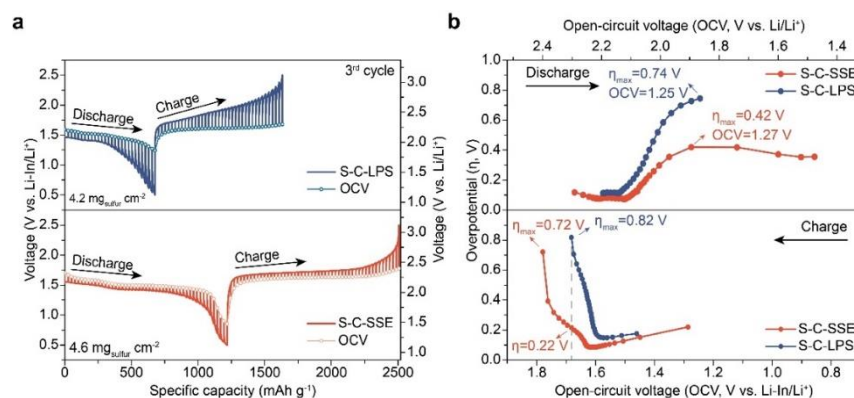
To construct efficient electronic conductive network, we have screened various commercial carbon additives, including Super-C carbon, Ketjen Black EC-600J (KB), carbon nanotube, carbon nanofiber and many other carbon materials. We found that sulfur cathode utilizing KB achieved best sulfur utilization and specific capacity. We have also investigated different methods to prepare carbon-sulfur composite and tested the impacts on cathode performance. After optimizing the cathode preparation process and conditions, sulfur cathode utilizing KB carbon materials demonstrated best performance. The specific capacity of sulfur cathode could reach over  $1470 \text{ mAh g}^{-1}$  at  $0.1 \text{ C}$ , over  $1000 \text{ mAh g}^{-1}$  at  $0.3 \text{ C}$  and even over  $880 \text{ mAh g}^{-1}$  at  $0.5 \text{ C}$  with sulfur loading of over  $2 \text{ mg cm}^{-2}$  and sulfur content of  $50 \text{ wt\%}$  at  $60 \text{ }^\circ\text{C}$ .

We found that the new solid electrolyte LPB we prepared could effectively improve the performance of high-sulfur-content cathodes ( $60\text{wt\%}$  of sulfur). To explicate sulfur cathodes' electrochemical behaviors during charge/discharge, we performed the galvanostatic intermittent titration technique (GITT) using Li-S ASSBs with sulfur cathodes using conventional  $75\text{Li}_2\text{S}-25\text{P}_2\text{S}_5$  (LPS) solid electrolyte (S-C-LPS) and the new solid electrolyte we prepared (S-C-SSE).



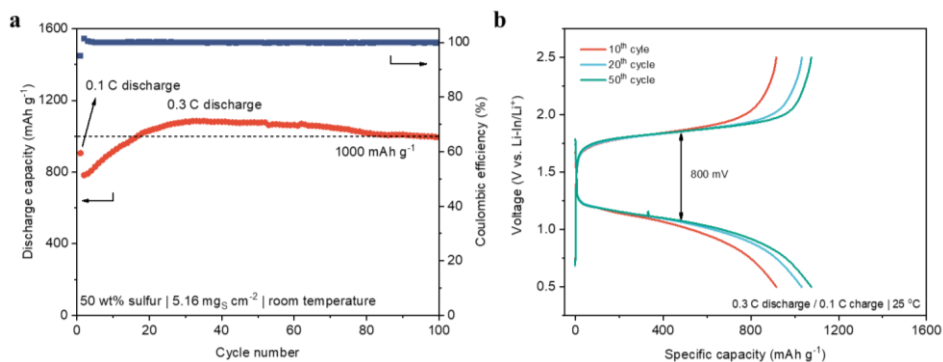
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As shown in Figure 5, S-C-SSE cathode delivered significantly higher specific capacity and lower overpotential than S-C-LPS cathode, suggesting more active sulfur, lower reaction resistance and improved  $\text{Li}^+$  transport kinetics in the S-C-SSE cathode than in the S-C-LPS cathode. Together, the analysis demonstrates that sufficient  $\text{Li}^+$  transport pathways and fast  $\text{Li}^+$  transport kinetics are achieved in the S-C-SSE cathode.



**Figure 5.** (a) GITT and open circuit voltage (OCV) curves of S-C-SSE and S-C-LPS cathodes at the third cycle. Current pulses of 0.03 C for 30 mins were employed, followed by 4-hour resting. (b) Overpotential profiles of S-C-SSE and S-C-LPS cathodes from the GITT measurement.

We continued working on optimizing sulfur cathodes in terms of compositions and preparation process. The new SSE we developed previously with high ionic conductivity of  $\sim 4 \text{ mS cm}^{-1}$  was used for sulfur cathode preparation. As a result, the sulfur cathode with optimized compositions, high sulfur content of  $\geq 50 \text{ wt}\%$ , high areal sulfur loading of  $5.16 \text{ mg}_S \text{ cm}^{-2}$  and preparation procedure delivered initial discharge capacity of  $905.3 \text{ mAh g}^{-1}$  at 0.1 C ( $1 \text{ C} = 1675 \text{ mAh g}^{-1}$ ). Upon increasing the discharge current rate to 0.3 C, the discharge capacity first decreased to  $\sim 782.8 \text{ mAh g}^{-1}$  then gradually increased to over  $1000 \text{ mAh g}^{-1}$  in 16 cycles (Figure 6a). The highest discharge capacity reached  $\sim 1085 \text{ mAh g}^{-1}$ . After 100 cycles, the discharge capacity remained  $993.4 \text{ mAh g}^{-1}$ . The performance meets the requirements for the milestone of the third quarter. As shown in the galvanostatic charge-discharge profiles (Figure 6b), the voltage polarization is  $\sim 800 \text{ mV}$  which is quite high. It indicates that ionic and electron transport in sulfur cathodes need further improvement toward better electrochemical performance of Li-S all-solid-state batteries with high areal sulfur loading at room temperature.



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**Figure 6. (a)** Cycling performance and **(b)** corresponding galvanostatic charge-discharge profiles of sulfur cathodes using the new SSE we developed at room temperature.

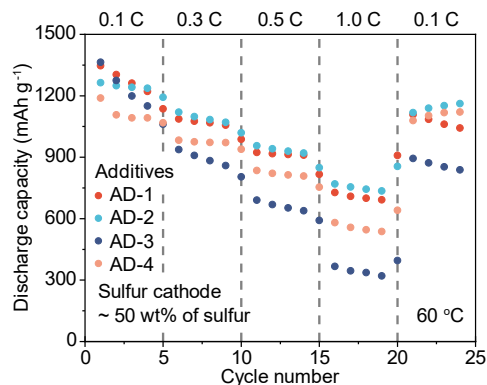
### 3) Solid additives for sulfur cathode

We have found that electrochemically active molecule (EAM) solid additives could impact charge/discharge behavior and improve cathode performance. To identify potential solid additives for sulfur cathode and investigate their impacts, we worked on both inorganic and organic solid additives.

For inorganic solid additives, we picked  $\text{TiS}_2$  for our study due to its special electronic conductive properties and lithiation/delithiation feature upon cycling. It was found that with  $\text{TiS}_2$  addition ( $\sim 0.38$  wt% in the cathode) to replace solid electrolyte (LPS), rate performance and sulfur utilization of sulfur cathode was improved. At 0.1 C, specific capacity of sulfur cathode could be increased from around  $1200 \text{ mAh g}^{-1}$  to  $1400 \text{ mAh g}^{-1}$  with the help of  $\text{TiS}_2$  addition. The sulfur content of the cathode is around 39 wt% and the test was performed at  $60^\circ\text{C}$  using LPS glass solid electrolyte.

For organic solid additives, we have identified an EAM solid additive which could significantly increase sulfur cathode performance. By using such EAM solid additive, we have demonstrated sulfur cathode (sulfur content = 50 wt%,  $1.5 \text{ mg}_{\text{Sulfur}} \text{ cm}^{-2}$ ) adding only 1 wt% EAM with a initial capacity of around  $1400 \text{ mAh g}^{-1}$  at 0.3 C. After 50 cycles, the specific capacity of sulfur cathode could still retain at above  $1200 \text{ mAh g}^{-1}$  at 0.3 C. LPS glass solid electrolyte was used for testing. The cycling of the cell was conducted at  $60^\circ\text{C}$ .

We further continued the screening of various cathode additives aiming to promote the interfacial ion transport. Four different cathode additives were selected and evaluated in sulfur cathodes with 50wt% of sulfur and areal sulfur loading of  $2\text{-}3 \text{ mg cm}^{-2}$ . As shown in Figure 7 while all additives can help promote initial discharge capacity to above  $1000 \text{ mAh g}^{-1}$  at 0.1 C, AD-2 additive enabled sulfur cathodes with the best rate performance and reversibility. Such improvement might be caused by the improved robust interfacial contact as well as faster interfacial ion transport between sulfur and solid-state electrolyte (SSE) due to the use of additives.

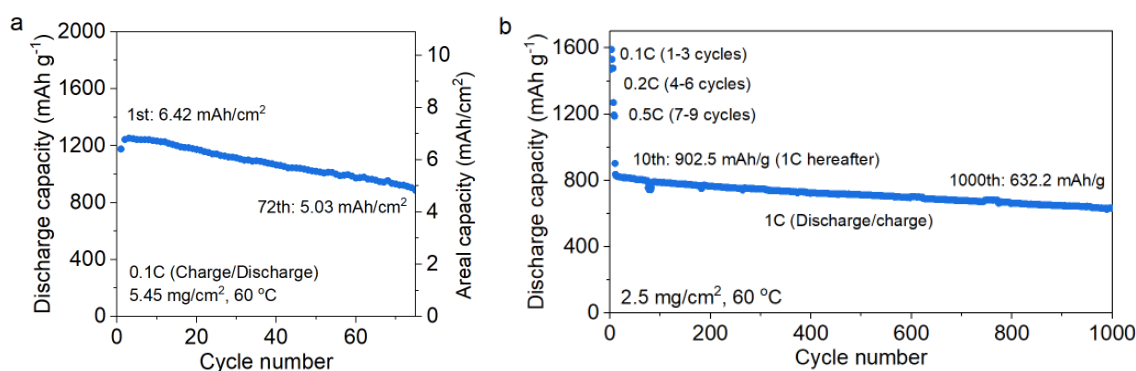


**Figure 7.** Rate performance of sulfur cathodes using different cathode additives at  $60^\circ\text{C}$  with an areal sulfur loading of  $2\text{-}3 \text{ mg cm}^{-2}$ .

### 4) Optimized solid-state cell

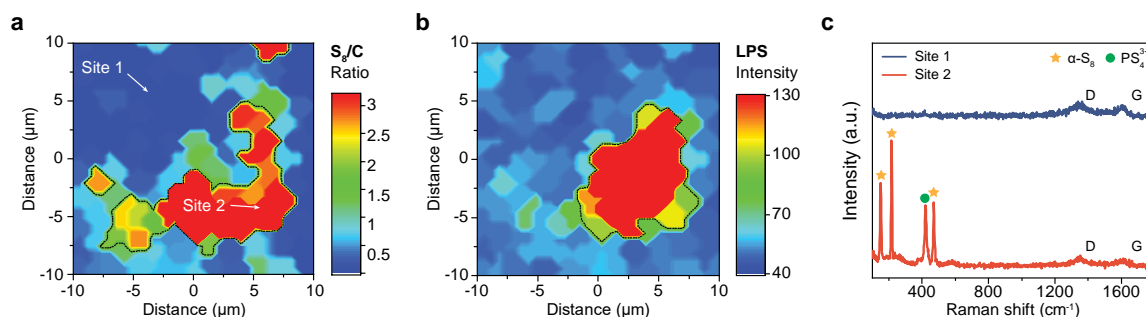
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To achieve a high loading cell with over  $5 \text{ mg cm}^{-2}$  and high discharge capacity of over  $1000 \text{ mAh g}^{-1}$  at  $0.3 \text{ C}$  rate, all-solid-state Li alloy anodes were also studied because of the high theoretical capacity of Li alloy anodes and their potential of suppressing dendrites. The alloy anodes we prepared have higher energy density than conventional Li-In alloy anode and improved cycling stability. The optimized lithium alloy with better lithium diffusion and lower impedance enables a superior electrochemical performance. As shown in Figure 8a, a cell composed of lithium alloy anode and high loading sulfur cathode ( $5.45 \text{ mg cm}^{-2}$ ) achieved an initial capacity of  $1176.9 \text{ mAh g}^{-1}$  at  $0.1 \text{ C}$  rate and  $60 \text{ }^\circ\text{C}$ . It can maintain above  $5 \text{ mAh cm}^{-2}$  until  $72^{\text{th}}$  cycle. Moreover, the optimized lithium alloy anode paired with normal capacity loading around  $2.5 \text{ mg cm}^{-2}$  was also fabricated and tested under step-increased currents from  $0.1\text{C}$ ,  $0.2\text{C}$ , and  $0.5\text{C}$  after every 3 cycles and then  $1\text{C}$  hereafter (Figure 8b). The long-term cycling test under  $1\text{C}$  shows stable discharge capacity retention over 1000 cycles. The results imply that the cell performance is promising and possible to replace conventional Li-In alloy anode.



**Figure 8.** (a) Cycling performance of the high loading cell with  $5.45 \text{ mg cm}^{-2}$  sulfur cathode using the lithium alloy anode under  $0.1\text{C}$ , (b) cycling performance of the cell with  $2.5 \text{ mg cm}^{-2}$  sulfur loading tested under  $1\text{C}$  rate for over 1000 cycles.

As the two-electron conversion reaction between  $\text{S}_8$  and  $\text{Li}_2\text{S}$  involves the formation of  $\text{Li}_2\text{S}_2$  as an intermediate state, it is not clear whether the low sulfur utilization is induced by unreacted sulfur or insufficient conversion of  $\text{S}_8$  to  $\text{Li}_2\text{S}$ . Therefore, we carried out Raman spectral imaging characterization of lithiated sulfur cathodes to understand the low sulfur utilization in conventional sulfur cathodes with a sulfur content of 50 wt% and  $75\text{Li}_2\text{S} \cdot 25\text{P}_2\text{S}_5$  (LPS) SSE. As shown in Figure 9, elemental sulfur with several micrometers in size can be clearly observed on the surface of the lithiated sulfur cathode. Because of the low electronic and ionic conductivity of sulfur, the sulfur, which is isolated from electronic conductive carbon framework (carbon) or ionic conductive carbon framework, cannot be reduced and thus induce the low sulfur utilization of the cell.



**Figure 9.** Raman spectral mapping images ( $10 \times 10 \mu\text{m}$ ) of the sulfur electrode surface after first discharge at 0.1 C and 60 °C showing the distribution of sulfur/carbon ratio (based on intensity) (a) and LPS SSE (b). (c) Raman spectra of the sulfur cathode surface at two different sites.

## Conclusions

In conclusion, we have accomplished our goals for the budget period 1-3 and obtained the following main achievements: (1) We successfully synthesized an effective additive for sulfur cathodes and obtained superior electrochemical performance. (2) The effects of ion/electron transport within sulfur cathodes were investigated through GITT, Raman technique, and XPS analysis. (3) We developed advanced sulfur cathodes for all-solid-state batteries with high sulfur content of  $\geq 50$  wt%, high areal sulfur loading of  $5.16 \text{ mgs cm}^{-2}$  which could deliver initial discharge capacity of  $905.3 \text{ mAh g}^{-1}$  at 0.1 C and maintain high discharge capacity at 0.3 C for over 100 cycles. (4) Alloy anodes were utilized to facilitate Li-S all-solid-state batteries, which performed stable cycling over 200 cycles, and showed good performance at high loadings or under high current conditions.

## Products and Technology

### Publications

1. Wang, D., Jhang, L.J., Kou, R. *et al.* Realizing high-capacity all-solid-state lithium-sulfur batteries using a low-density inorganic solid-state electrolyte. *Nature Communication* **14**, 1895 (2023).