Supplementary Information

Electrolyte Strategies for Practically Viable All-Solid-State Lithium-Sulfur Batteries

*Gordon Jarrold and Arumugam Manthiram**
Materials Science and Engineering Program and Walker Department of Mechanical Engineering
The University of Texas at Austin
Austin, TX 78712, USA
E-mail: manth@austin.utexas.edu

Supplementary Table 1: Gravimetric densities of various SSEs and cell component materials

The following materials parameters were used for SSE cathode volume calculations and energy density calculations:

Material	ρ (g cm-3)[a]	Refs
Lithium	0.53	-
LPSC	1.64	MP
LGPS	2.04	MP
$Li_7P_3S_{11}$	1.87	MP
Li_3PS_4	1.83	MP
LLZO	5.1	MP
LLTO	5	[1]
LATP	2.92	[2]
LIC	2.59	MP
LYB	3.54	MP
$LiBH_4$	0.7	MP
Carbon	1.9	-
Sulfur	2.07	-
Copper	8.9	-
Aluminum	2.7	-

[a]SSE densities were taken from Materials Project (MP) unless noted otherwise.

Supplementary Table 2: Ionic conductivities of various SSEs

Material	σ (mS cm^{-1})	Refs
LPSC	1 – 5	[3]
$LPSC_{1.5}$	3 – 9	[4]
LGPS	1 – 12	[5]
$Li_7P_3S_{11}$	1 – 17	[6]
Li_3PS_4	0.1 – 0.5	[7]
LLZO	0.1 – 1	[8]
LLTO	0.1 – 1	[9]
LATP	0.3 – 1	[10]
LIC	1 – 2	[11]
LYB	1 – 4	[12]
$LiBH_4$	1e-5 – 1e-4	[13]

Supplementary Methods:

SSE volume fraction analysis

To calculate the volume fraction of SSE in a 30 : 50 : 20 mass ratio cathode, the following equation was used:

$$V_{cathode} = \frac{m_{sulfur}}{\rho_{sulfur}} + \frac{m_{carbon}}{\rho_{carbon}} + \frac{m_{SSE}}{\rho_{SSE}}$$

A total cathode mass of 1 g was assumed for simplicity. The same equation was used to calculate the mass fraction of SSE needed to obtain a cathode with 50 volume % of SSE, with the assumption that the S / C ratio remained 3 / 2. To calculate the cathode gravimetric energy density, the following equation was used:

$$Cathode\ Gravimetric\ Energy\ Density = \frac{m_{sulfur} \times 1{,}672\ mA\ h\ g^{-1} \times 0.85 \times 1.85\ V}{m_{sulfur} + m_{carbon} + m_{SSE}}$$

A total cathode mass of 1 g was assumed for simplicity. A sulfur utilization of 85% and an average discharge voltage of 1.85 V are assumed.

Cell-level gravimetric energy density

To calculate the cell-level gravimetric energy densities, all the capacity was assumed to come from sulfur at 85% sulfur utilization at an average discharge voltage of 1.85 V. For cells with an $InLi_{0.5}$ anode, the average discharge potential was reduced to 1.23 V because $InLi_{0.5}$ operates at 0.62 V vs. Li/Li^+. No capacity was assumed to come from SSE redox. The cell area was assumed to me 1 cm^{-2} for simplicity. An N/P ratio of 2 was used for Li-metal, while a mass of 300 mg was assumed for $InLi_{0.5}$ anodes (N/P ≈ 10 for a 2 mg sulfur cathode). A separator mass of 120 mg was assumed for thick separators and 6 – 10 mg for 30 µm – 50 µm separators. The 30 % sulfur content cathode was assumed to follow a 30 : 50 : 20 mass ratio, while the 50 % sulfur content cathode was assume to follow a 50 : 35 : 15 mass ratio for sulfur : SSE : carbon. Copper and aluminum current collectors were assumed to be 10 µm thick on either side of the cell stack. The following equation was used:

$$Gravimetric\ Energy\ Density = \frac{m_{sulfur} \times 1{,}672\ mA\ h\ g^{-1} \times 0.85 \times 1.85\ V}{m_{cathode} + m_{SSE_{separator}} + m_{anode} + m_{current\_collectors}}$$

The mass of Li metal was calculated with the following equation:

$$m_{Li-metal} = m_{sulfur} \times \frac{N}{P} ratio \times \frac{1{,}672\ mA\ h\ g^{-1}}{3{,}860\ mA\ h\ g^{-1}}$$

Supplementary References

1. Mena, M. F. *et al.* Electrochemical performance enhancement of perovskite-type Li0.3La0.57TiO3 ceramic electrolyte by controlling synthesis parameters. *J. Eur. Ceram. Soc.* **45**, (2025).
2. Waetzig, K. *et al.* Synthesis and sintering of Li1.3Al0.3Ti1.7(PO4)3 (LATP) electrolyte for ceramics with improved Li+ conductivity. *J. Alloys. Compd.* **818**, (2020).
3. Boulineau, S., Courty, M., Tarascon, J. M. & Viallet, V. Mechanochemical synthesis of Li-argyrodite Li_6PS_5X (X = Cl, Br, I) as sulfur-based solid electrolytes for all solid state batteries application. *Solid State Ion.* **221**, 1–5 (2012).
4. Adeli, P. et al. Boosting solid-state diffusivity and conductivity in lithium superionic argyrodites by halide substitution. *Angew. Chem. Int. Ed. Engl.* **58**, 8681–8686 (2019).
5. Tao, B. et al. Thio-/LISICON and LGPS-type solid electrolytes for all-solid-state lithium-ion batteries. *Adv. Funct. Mater*. **32** (2022).
6. Chang, D., Oh, K., Kim, S. J. & Kang, K. Super-ionic conduction in solid-state Li7P3S11-type sulfide electrolytes. *Chem. Mater.* **30**, 8764–8770 (2018).
7. Yamamoto, K. et al. High ionic conductivity of liquid-phase-synthesized Li3PS4 solid electrolyte, comparable to that obtained via ball milling. *ACS Appl. Energy Mater.* 4, 2275–2281 (2021).
8. Sastre, J. et al. Lithium garnet Li7La3Zr2O12 electrolyte for all-solid-state batteries: closing the gap between bulk and thin film Li-ion conductivities. *Adv. Mater. Interfaces* 7, 2000425 (2020).
9. Wang, M. J., Wolfenstine, J. B. & Sakamoto, J. Mixed Electronic and Ionic Conduction Properties of Lithium Lanthanum Titanate. *Adv Funct Mater* **30**, (2020).

10. C. Luo, M. Yi, Z. Cao, W. Hui, Y. Wang. Review of ionic conductivity properties of NASICON type inorganic solid electrolyte LATP. *ACS Appl. Electron. Mater*., **6**, 641 (2024)

11. Wen, S., Sheng, H., Zhang, Y., Zheng, S. & Wang, Z. The effect of HCl on achieving pure Li3InCl6 by water-mediated synthesis process. *J. Alloys. Compd.* **984**, (2024).

12. Poudel, T. P. *et al.* Sliceable, Moldable, and Highly Conductive Electrolytes for All-Solid-State Batteries. *ACS Energy Lett.* **10**, 40–47 (2025).

13. Gulino, V., Barberis, L., Ngene, P., Baricco, M. & De Jongh, P. E. Enhancing Li-Ion Conductivity in LiBH4-Based Solid Electrolytes by Adding Various Nanosized Oxides. *ACS Appl Energy Mater* **3**, 4941–4948 (2020).