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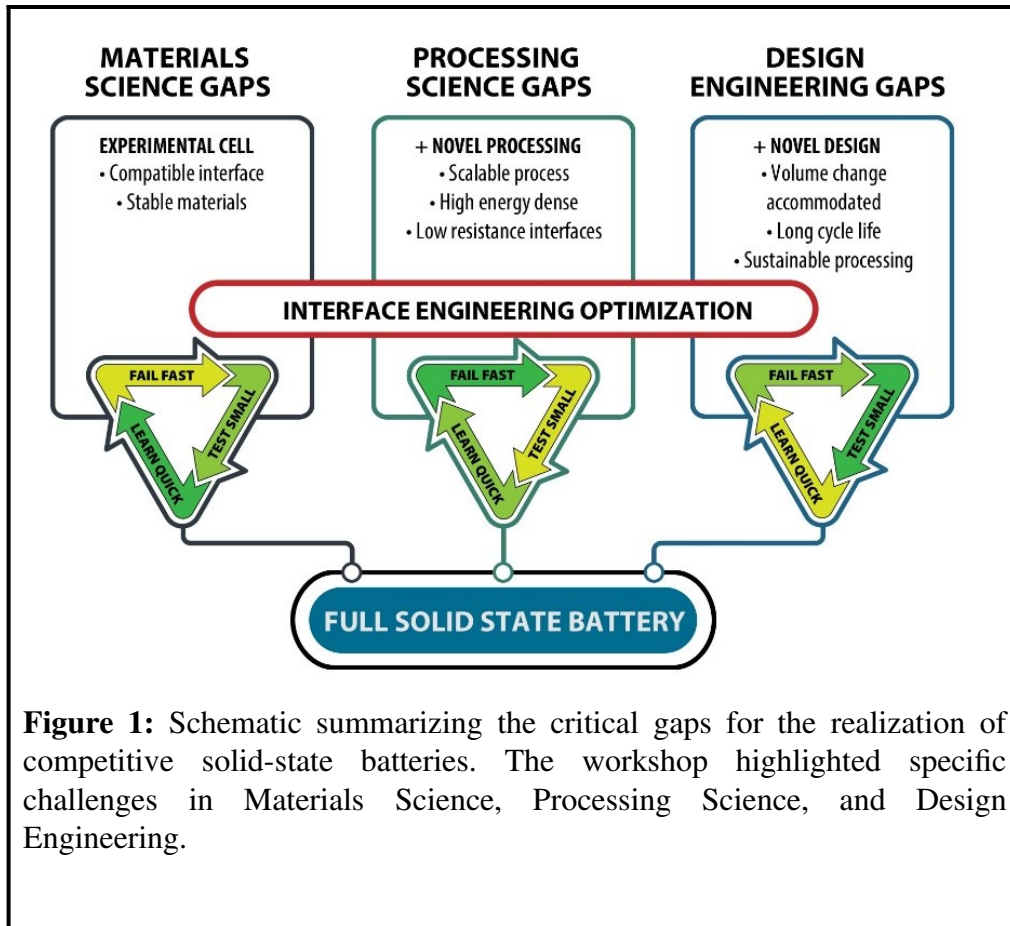
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CHALLENGES FOR AND PATHWAYS TOWARD LI-METAL BASED ALL SOLID-STATE BATTERIES

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Solid-state batteries utilizing Li metal anodes have the potential to enable improved performance (specific energy >500 Wh/kg, energy density >1,500 Wh/L), safety, recyclability, and potentially lower cost (< \$100/kWh) compared to advanced Li-ion systems.^{1, 2} These improvements are critical for the widespread adoption of electric vehicles and trucks and could create a short haul electric aviation industry.¹⁻³ Expectations for solid-state batteries are high, but there are significant materials and processing challenges to overcome.

On May 15th, 2020, Oak Ridge National Laboratory (ORNL) hosted a 6-hour, national online workshop to discuss recent advances and prominent obstacles to realizing solid-state Li metal batteries. The workshop included more than 30 experts from national laboratories, universities, and companies, all of whom have worked on solid-state batteries for multiple years. The participants' consensus is that, although recent progress on solid-state batteries is exciting, much has yet to be researched, discovered, scaled, and developed. Our goal was to examine the issues and identify the most pressing needs and most significant opportunities. The organizers asked workshop participants to present their views by articulating fundamental knowledge gaps for materials and processing science, mechanical behavior and battery architectures critical to advancing solid-state battery technology. The organizers used this input to set the workshop agenda. The group also considered what would incentivize the adoption of US manufacturing and how to accelerate and focus research attention for the benefit of the US energy, climate, and economic interests. The participants identified pros and cons for sulfide, oxide, and polymer-based solid-state batteries and identified common science gaps among the different chemistries. Addressing these common science gaps may reveal the most promising systems to pursue in the future.



A comprehensive document was drafted and published as an ORNL technical document (doi:10.2172/1731043).⁴ The document reports a consensus of the most essential considerations to enable low-cost, safe, high-performance, long-lasting, and scalable solid-state batteries. As shown in Figure 1, this focus article summarizes the following main findings of the workshop (see the ORNL technical document for more details):

- I. Materials Science Gaps
- II. Processing Science Gaps
- III. Design Engineering Gaps

Although not stated as specific goals for the workshop, participants also addressed practical tradeoffs in manufacturing processes and efficiency, materials costs and handling, and environmental sensitivity. Discussions touched on opportunities and barriers for domestic battery manufacturing. Participants agreed that testing standardization and statistical analysis of solid-state battery performance are critical to advance the field.⁵ Currently, reported properties and cell performance vary unacceptably among laboratories studying nominally the same materials. A careful safety evaluation is also needed to quantify solid-state battery safety versus that of leading Li-ion designs, including the important issue of whether a small amount of liquid may be added to improve performance without compromising safety and other benefits.

To complement the workshop discussion and assess state-of-the-art developments, the organizers performed a literature analysis of solid-state batteries. Figure 2a shows the number of peer-reviewed publications over the timespan 2000-2020. The volume of literature and rate of publication has increased significantly over the last decade. To ensure a representative perspective, a dozen recent review articles were analyzed based on their emphasis on key technical areas for solid-state battery development (see Figure 2b).⁵⁻¹⁶ The analysis revealed that researchers have made significant progress in new materials discovery, but integrating these materials into practical devices has lagged. The dearth of relevant prototype cell data may be due to an underemphasis on processing science and solid-state mechanics, as well as the challenges for the single-PI research model to overcome the challenges in producing high-quality prototype cells. This analysis is consistent with the discussions held during the workshop.

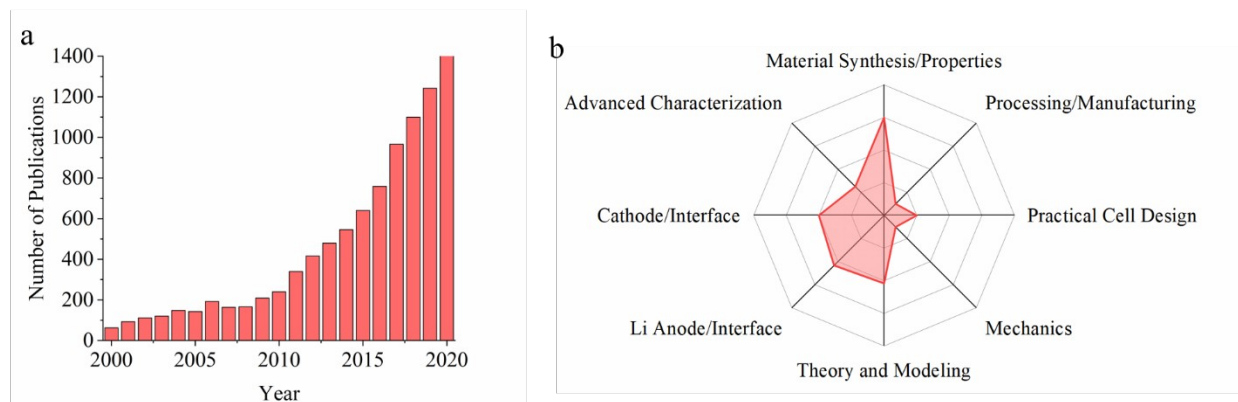


Figure 2. Solid-state battery literature analysis showing (a) peer-reviewed publications from 2000 to 2020 (keywords: lithium and solid-state battery*, Web of Science) and (b) a radar plot that compares the level of activities in key technical areas for solid-state batteries based on analysis of 12 recent review articles.⁵⁻¹⁶

I. MATERIALS SCIENCE GAPS

Progress on solid-state batteries surges following the discovery of promising solid electrolytes. However, every known solid electrolyte has one or more drawbacks that must be overcome to enable the development of viable solid-state batteries for EVs. Work should continue to discover new electrolytes, with the expectation that other performance and processing criteria are simultaneously satisfied. Furthermore, a clear understanding of the challenges to integrating components into batteries will inform the search for new materials.

I.1 Science Gaps for the Li metal anode

The Li metal anode is common to all the batteries considered at the workshop, yet this component may be the least studied. Li metal has recently captured more attention from the US Department of Energy (DOE) Vehicle Technologies Office and the Advanced Research Projects Agency-Energy (ARPA-E). There was considerable discussion among participants on this topic. “We know so much more now than just 5 years ago, but we are just getting started,” reported Paul Albertus from the University of Maryland. One key finding, by nanoindentation and

compression of Li micropillars, is that when the volume of Li is small, the hardness and yield strength can be much larger than that of bulk Li.^{17, 18} Consequently, we need to determine the relevant length scale for mechanical tests to inform our understanding of the Li anode and the mechanisms leading to Li redistribution, particularly when related to battery failure

The following questions need to be answered to fill the science gaps that exist in the development of an optimized Li metal anode:

- What defect generation/annihilation processes operate in Li films (< 30 μm thick) when Li is plated and stripped through a generic solid electrolyte?
- What conditions (e.g., rate, temperature, applied stress, and duty cycle history) modify Li plating and stripping behavior?
- What are the stress relaxation mechanisms for Li, and how do they change with the type and magnitude of the stress field, the mechanical boundary conditions, and the strain rate?
- How do defects such as grain boundaries, dislocation density, elemental impurities, and alloying elements alter the properties and cycling performance of Li metal anodes?
- Is a Li seed layer needed to template plated Li or provide mechanical compliance to improve cycling stability?
- How do interphase regions, formed by reactions or additions at the Li/solid electrolyte interface, govern transport?

I.2 Science Gaps for the Solid Electrolyte in Contact with Li Metal

The community has learned much about failure at the Li/solid electrolyte interface in recent years. More specifically, we see that (1) effective passivation of the interface reduces Li consumption, (2) a high modulus solid electrolyte formed with a dense, smooth interface suffers fewer issues related to Li roughening, (3) a higher fracture toughness inhibits cracks that may form shorts, and (4) higher electronic resistivity mitigates Li^+ reduction within the solid electrolyte separator. Given this background, several important questions were identified:

- What promotes electrochemical stability or kinetically-limited passivation with Li?
- What mechanisms are available to strengthen solid electrolyte properties at the appropriate length scale, improve stability, and inhibit failures/fatigue during extended Li cycling?
- How do the bulk properties of the solid electrolyte and its surface chemistry/homogeneity (e.g., current uniformity) affect Li cycling?

- How does the cathode influence the Li anode interface during cell cycling?

I.3. Science Gaps for Active Cathode Materials and Solid-State Composite Cathodes

For the highest energy density, the cathode must be the most voluminous component of the battery. For example, suppose the cathode serves as the mechanical support and battery substrate. In that case, the current collectors, separator, and Li anode can all be applied as thin coatings with limited volume, weight, and cost as shown in Figure 3. In traditional slurry cast cathodes, organic binders may suffice to form freestanding cathodes, or polymer electrolytes can be added to fill voids and facilitate Li^+ transport. The composite cathode may also be bonded, fused, or sintered to improve interfacial contact. These steps complicate the processing but ensure the formation of mechanically robust solid-solid interfaces. The key is to fabricate a cathode that will (i) withstand dynamic stresses during cycling and (ii) provide facile electronic and ionic transport at low stack pressure (<1 MPa).

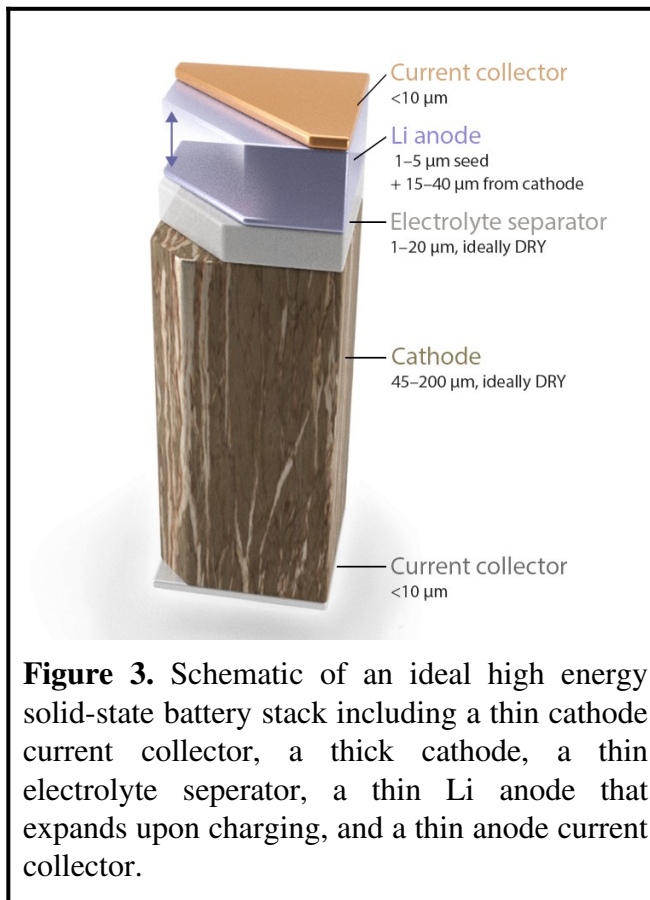


Figure 3. Schematic of an ideal high energy solid-state battery stack including a thin cathode current collector, a thick cathode, a thin electrolyte separator, a thin Li anode that expands upon charging, and a thin anode current collector.

One viable approach for solid-state cathodes is the use of composites where various components are added to manage the bonding and transport. The added phases may be a solid catholyte, an electronic conductor, or a solid mixed ionic-electronic conductor. The interfaces among these phases must be electrochemically stable, have low bulk and grain/particle boundary resistance for ion and charge transport, and survive repeated volume changes of the active material. Cathodes with reasonable cycling have been demonstrated with oxide or phosphate active materials (LiCoO_2 [LCO], $\text{LiNi}_x\text{Mn}_y\text{Co}_{1-x-y}\text{O}_2$ [NMC], LiFePO_4 [LFP]) mixed with carbons and either lithium thiophosphate or polymer electrolytes. Generally, cathode particles embedded in a soft electrolyte function better when: (i) stack pressure is applied to preserve interfacial contacts and/or (ii) cycled at elevated temperature which improves transport rates and reaction kinetics.

Participants offered comments on their experience and expectations for promising cathode development. Waxy plastic crystal electrolytes represents one class of soft catholytes able to accommodate strain associated with cathode volume changes.¹⁹ Similar success has been demonstrated for slurry cathodes filling a 3D porous $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) sintered template.¹⁹ An interesting alternative is to identify and process soft organic cathode materials that provide

good contact with hard solid electrolytes. How thick cathodes impact volume changes at the full cell level is still unexplored. Researchers and industry developing practical sulfide-based solid-state batteries find that appreciable external stack pressure (often on the order of 10 MPa) is needed to maintain contacts during cycling, but these experimental housings are massive. Discussion ventured to the possibility of cleverly designed composites in which internal pressure develop during cycling to maintain interfacial contacts. Below we pose some key scientific challenges related to this topic:

- What can be achieved through defect and microstructure engineering to enhance reaction kinetics and mechanical properties for dense, single-phase cathodes at all states of charge?
- How can texture and grain structure be used to improve reaction kinetics and mitigate solid electrolyte fracture?
- What are the relative pressure and electrochemical driving forces experienced by the active cathode material? Is there a restoring component that can provide a more homogeneous reaction?
- What cathode design principles will lead to a resilient cathode-catholyte interface which maintains intimate contact during cycling?

II. PROCESSING SCIENCE GAPS

Advanced processing methods create opportunities for new and modified materials that are not available with conventional approaches. While many challenges related to materials and interfaces remain unsolved, an effort to understand processing hurdles may save significant time and effort. Advanced materials processing could also open new directions for solid-state batteries or accelerate the development of current materials.

The following examples illustrate how unique processing approaches can be used to form intimate contact between the cathode and solid electrolyte. Sintering is often required to form good interfacial contact between the cathode and the solid electrolyte when simple cold pressing is inadequate, especially for oxide solid electrolytes. Bonding agents, so long as they do not compromise electrochemical and mechanical properties, are useful to densify and fuse the active material and solid electrolyte phases at lower temperatures. Studies of rapid thermal annealing (e.g., radiative heating from a carbon ribbon²⁰) may open new and practical processing directions. Interfaces with uniform contact may also be achieved by filling or coating the interface with a liquid that is later solidified. For example, a porous 3D cathode can be back-filled with liquid precursors which are thermally treated to form a solid ionogel. There are also opportunities to investigate other novel and solvent-free processing routes to form intimate interfacial contact.

Processing determines the solid electrolyte separator microstructure and mechanical properties. There are well-known methods (e.g., precipitation hardening, phase transformation toughening, and tempering) to strengthen structural ceramic and glass materials, but similar mechanisms have not been reported for solid electrolytes. One processing science gap for solid-state batteries is

determining whether or not mechanisms exist to strengthen thin solid electrolytes and thick cathodes without impeding transport. Approaches that avoid inactive components are particularly attractive to maintain high specific energy. Advantages of glass and amorphous electrolytes include good chemical stability and ductility in addition to providing a naturally smooth surface. New insights are needed to process thin amorphous materials efficiently. We note that battery components are processed ideally as integrated coatings or composites, not as stand-alone parts.

Participants also discussed the connection between processing innovation and potential competitiveness of the US battery industry. There is tension between short-term commercialization with existing equipment and a long-term horizon employing an investment in a totally new processing line. Initial applications that are smaller and less demanding than EVs may provide important markets for solid-state batteries. All participants recognized that for commercialization, solid-state batteries must compete well on performance compared to advanced Li-ion technology, even if not at first on the price.

III. DESIGN ENGINEERING GAPS

Most solid-state cells are, at present, a stacked design with the cathode composite borrowed from a typical Li-ion architecture. Exceptions are the 3D designs formed using 3D-templated cathodes, or more recently, 3D-porous solid electrolytes formed by freeze casting or burnout of sacrificial components. Another is a so-called “2.5D” design consisting of a 2D Li-anode sheet with a 3D composite cathode. These designs have been adopted to increase the interfacial area and reduce local current density at the electrode-electrolyte contacts. Such approaches are promising, but in most cases, there is not a clear pathway to cost-effective scale-up. Alternative architectures may also enable mechanically robust structures and interfaces. Research in such structures is motivated by concerns that maintaining high stack pressure (≥ 5 MPa) on solid-state cells may require an external mechanical fixture that negates any specific energy/energy density benefit of cells. Discussion focused on how materials and composites should be processed and bonded for solid-state cathodes. Advanced manufacturing may make a variety of well-controlled architectures possible. Many interesting questions on this topic should be considered:

- Can volume changes be cleverly directed to provide internal compression at the material interfaces to replace large external stack pressures?
- Are there other ways to temper the cathode and its internal interfaces to strengthen and resist fracture?
- How should composites be designed to maintain their structure and internal connectivity during cycling?
- How do volume changes during cycling affect solid-state battery performance and packaging requirements?
- How can chemical reactions between the cathode and solid electrolyte during densification and cycling be mitigated?

IV PATHWAYS TO SOLID-STATE BATTERIES—ADDRESSING SCIENCE GAPS

IV.1 Control and Efficient Cycling of the Li Metal Anode

Sections I.1-2 highlight the science gaps and challenges associated with Li metal anodes. While the reasons for poor Li cycling with solid electrolytes are becoming clearer, the solutions are not. Applying large stack pressure is not a viable solution; if unavoidable, such an approach may be a showstopper for solid-state batteries in EVs. Issues that will inform understanding of the Li metal anode include research to (i) compare the Li cycling performance of full cells versus Li/Li cells, (ii) compare cycling performance at different temperatures, including where Li is molten, (iii) assess the effects of various impurities in the Li, and (iv) compare Li from different sources. For example, the Li sources may include commercially rolled and passivated ultrathin Li, vacuum-grown Li films, and Li grown electrochemically in an anode-free cell configuration. Alternatively, the option to use Li alloy anodes or to provide a scaffold for Li plating (e.g., a nano Ag-carbon composite, as reported by Samsung²¹) should not be ignored. However, these solutions will sacrifice overall energy density. Furthermore, it cannot be overemphasized that the path to understanding is to use methods and cells designed to “fail fast” and “learn quickly.” For Li anode studies, this means that both the metallic Li and solid electrolyte separator should be very thin, with little excess capacity to either sustain side reactions or perpetuate interface roughening that will eventually lead to failure.

IV.2 Solid-State Cathodes and Composites

The solid-state cathode is key to enable performance that matches or exceeds that of Li-ion batteries. For a battery with the highest energy density, the active cathode should occupy the largest fraction of the battery. As such, the composite cathode should act as the physical support. This has been a critical roadblock for practical solid-state batteries. In most designs, the cathodes do not deliver sufficient energy at room temperature and under reasonable stack pressures. Hybrid designs using standard Li-ion battery cathodes with liquid or gel catholyte have been tested, but they were also limited by interfacial reactions and poor transport.

Scientifically, the solid-state cathode is also a foundational research topic because it requires solutions for many critical interface science challenges. For example, the effects volume changes, interface integrity, and phase connectivity for facile ion and electron transport need to be addressed to develop cathodes which provide the required current density and areal capacity (1-10 mA/cm² and >3 mAh/cm², respectively). Studies on the defect equilibria with state of charge, the performance of solid interfaces between different materials, stress and fatigue effects, and stress relaxation mechanisms will provide insights that can be applied to other interfaces in solid-state batteries. Research on solid-state cathodes must also address science gaps in materials, processing, and battery architecture.

For a successful solid-state cathode, experts in battery materials and solid-state electrochemistry should work closely with experts in materials mechanics and processing to (i) minimize and relieve cycling-induced stresses, (ii) identify mechanisms and architectures for strengthening the materials and interfaces, (iii) utilize materials and processes to form direct ion-transport paths that span the thickness of the cathode, and (iv) reduce or eliminate the need for external stack

pressure. As mentioned in the proposed Li studies, projects should implement procedures to “test small” and “fail fast” to advance scientific and technological knowledge of the solid cathode as quickly as possible. For the cathode, this fail-fast strategy encourages researchers to reveal the presence of any residual solvent, test at ambient temperature and pressure, and assess the cathode’s effective specific capacity which includes all components (e.g., active material, conductive additives, and catholyte). Progress can also be enhanced by studying model cathode interfaces, adopting aggressive cycling conditions, determining chemical/mechanical data for computational modeling, and developing advanced characterization tools to probe interfaces during and after cycling.

SUMMARY

Li metal solid-state batteries have the potential to provide advantages in energy density, safety, cost, and recycling over current state-of-the-art Li-ion systems. However, success is not assured, and solid-state battery development faces several challenges including (i) improving control of materials and interfaces, (ii) addressing processing challenges and cost, (iii) demonstrating performance which exceeds that of advanced Li-ion batteries, and (iv) maintaining optimal stack pressure for solid-state battery packs without affecting cost and energy density.

Delivering a full solid-state battery that meets the performance, cost, and manufacturability necessary for an electric vehicle within the next 5 years, or even 10 years, is ambitious, particularly when the best solid electrolyte and cathode chemistry are not obvious. Targeting a less-demanding application such as a consumer electronics battery may lead to short-term successes but risks diverting attention from the most challenging problems for large-scale EV and grid scale deployment. There are already strong scientific efforts underway that could be expanded to emphasize mechanical, transport, and cell-level measurements. It is worth creating efforts that include statistics and process control to facilitate reproducibility among different groups and institutions. Attention to battery failure mechanisms and large-scale, organized, and integrated data analysis should also be emphasized. Input from companies with tremendous expertise and knowledge organized around the practical aspects of making devices could significantly impact research progress.

In summary, solid-state batteries hold great promise for high energy batteries for EVs and other applications. While the potential is great, success is contingent on solving critical challenges in materials science, processing science, and fabrication of practical full cells. This article has outlined several key challenges in the hope that they will encourage and inspire solutions and the eventual realization of high energy solid-state batteries.

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