

Promising All-Solid-State Batteries for Future Electric Vehicles

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The global electric vehicle (EV) market has grown sharply over the past decade on the back of technological advances and supportive policies. There were about 7.2 million EVs on the world's roads in 2019, and this number is predicted to near 140 million within the next decade.¹ Among the various types of secondary batteries, lithium-ion batteries (LIBs) have been a key technology in the success of EVs owing to their high energy densities and long service lives. However, current LIBs are expensive, with limited ability to sufficiently deliver the performance required to advance EV development. In particular, the short driving range achievable with a single charge and long charging times are the main concerns of the broad consumer market.² To achieve EV performances comparable to those of internal combustion engine vehicles, many researchers and manufacturers are devoted to developing LIBs with higher power and energy densities.

However, this drive to develop more efficient and powerful LIBs has exposed the safety challenges of certain approaches. The tight packing of electrodes and their narrow separation increase the energy densities of LIBs but also increase their risk of explosion. The occasional reports of exploding or combusting LIBs in mobile phones, EVs, and energy storage systems are enough to make the average consumer nervous, possibly damaging market demand for EVs. Hence, safe LIBs are a prerequisite for the further acceleration of EV deployment. The liquid organic electrolyte (LE) is the component of an LIB primarily responsible for any combustion risk because of its high volatility and flammability. In this regard, all-solid-state batteries (ASSBs), in which solid electrolytes (SEs) are used as substitutes for LEs, are increasingly regarded as very promising next-generation battery systems.

In addition to being nonflammable, SEs have several advantages over conventional LEs. Current state-of-the-art LIBs have a narrow operating temperature range owing to the LE; at low temperatures ($< \sim 0$ °C) the ionic conductivity of the electrolyte is reduced and performance decreases significantly, whereas, at high temperatures ($> \sim 60$ °C), the high reactivity of LEs accelerates their decomposition and the deterioration of other components, thus causing battery swelling or malfunction.³ Unlike LEs, the intrinsic solid nature of SEs tolerates a wide operating temperature range (-30 to 100 °C) while delivering reliable performance.⁴ Moreover, the energy densities of ASSBs can be greatly increased by directly adopting lithium metal as an anode, as the solid–solid contact

between the electrolyte and lithium effectively disrupts the free growth of lithium dendrites, which are the cause of critical safety problems.^{5,6} In addition, SEs acting as rigid physical barriers between the anodes and cathodes enable bipolar electrode configurations, which is advantageous for optimizing energy density and utilizing limited space.

Despite several advantages that distinguish them from conventional LE-based batteries, considerable time and effort will be required before ASSBs are market-ready; the technology is still in an early research stage. First, high ionic conductivity, comparable to the level of LEs at operating temperature, is the most important criterion for efficient ASSBs because most SEs demonstrate inherently lower ionic conductivity than liquid electrolytes. Various organic and inorganic materials have been studied as possible substitutes for LEs, and recent research shows great potential. A number of oxide- and sulfide-based SEs have demonstrated high ionic conductivities near or even better than those of LEs (~ 1 – 10 mS cm⁻¹). For example, the oxide-based electrolyte, $\text{La}_{0.51}\text{Li}_{0.34}\text{TiO}_{2.94}$, and sulfide-based electrolytes, $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$ and $\text{Li}_{9.54}\text{Si}_{1.74}\text{P}_{1.44}\text{S}_{11.7}\text{Cl}_{0.3}$, achieved ionic conductivities of ~ 1 , 12, and 25 mS cm⁻¹, respectively.^{7–9} Given these advances, some argue that ionic conductivity, in itself, is not the bottleneck for high-performance ASSBs; however, creative solutions involving highly conductive SEs remain a challenge.

Second, reported ASSBs suffer from low Coulombic efficiency, poor power performance, and unstable cycling stability, largely resulting from high interfacial resistance at numerous solid–solid interfaces.^{10,11} Unlike LEs, SEs cannot completely cover the cathode or anode interfaces; consequent interfacial complications arise even at electrolyte–electrolyte interfaces despite the high pressures prevailing during cell assembly. As lithium ions can migrate only through intimate interfaces, the poor wettability of solid electrolytes with other components results in high charge-transfer resistance and thus the rapid deterioration of ASSB performance. The mechanical stresses induced by the continuous and repeated volume

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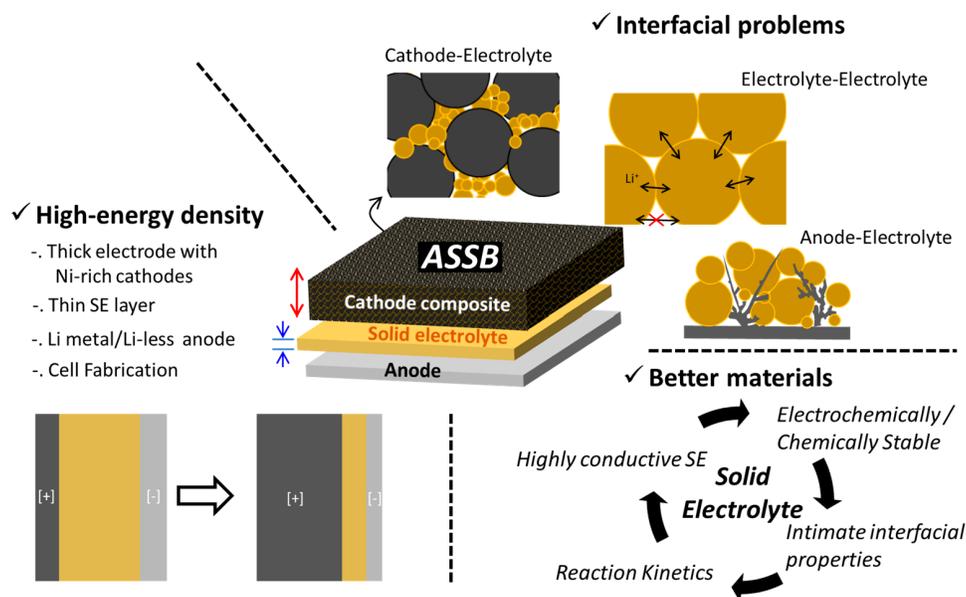


Figure 1. Technical challenges to ASSB optimization.

changes of the cathode and anode loosen contact, leading to the isolation of either active material.

Third, although it is generally believed that SEs exhibit a relatively wider electrochemical window, the practical range of the electrochemical stability window of SEs is another factor that adversely affects performance.^{10,11} Outside of the stable potential window, which depends on the environment between the active materials and SEs, spontaneous oxidative or reductive side reactions at the interfaces accumulate highly resistive solid electrolyte interphase (SEI) or cathode electrolyte interface (CEI) layers that further deteriorate Coulombic efficiency during cycling. Finally, some SEs are highly sensitive to ambient atmosphere, and byproducts resulting from atmospheric exposure can completely change the chemistry and kinetics at their interfaces; the consequent necessity of handling these materials under inert conditions is a serious impediment to associated development and research.¹²

ASSBs are promising options for next-generation battery systems. In order for ASSBs to be considered suitable candidates for replacing the current LIBs in EVs, the energy density and technical challenges must be resolved. The development of high-energy-density ASSBs requires advanced materials for optimum performance and a comprehensive understanding of the inner workings of the cells that is informed by various analytical tools. The design of highly conductive SEs should consider both chemical/electrochemical stabilities and interfacial mechano-electrochemistry.

In addition to novel SEs, research emphasis should be placed on the development of cathode and anode materials suitable for ASSB systems. Currently, Ni-rich layered oxide materials and lithium are leading candidates for cathodes and anodes, respectively, while lithium-free anodes are being explored as well.^{13,14} However, the interfacial stabilities, failure mechanisms, and mechano-electrochemistry of these ASSB components should be further addressed. Many interface limitations can be resolved through the effective interfacial engineering of cathode and anode materials; for example, a surface coating on active materials is a widely applied modification used to prevent side reactions while interfacial-engineered anode and

microstructure-tailored cathode materials remain in intimate contact with solid electrolytes by minimizing mechanical stresses during cycling.^{10,11,13,14} Their coupling with new SEs will enable ASSBs to accommodate high mass loadings of active materials and thin SE layers, resulting in higher energy densities, cycling efficiencies, and stabilities.

With this Editorial, we hope to stimulate interest in ASSBs and their technology. Extensive research (Figure 1) that delves into the basic mechanisms of this technology through multiscale analyses, insightful performance engineering, and practical fabrication is welcome, because the contributions of many researchers will be required to realize market-ready ASSBs.

Yang-Kook Sun, Senior Editor orcid.org/0000-0002-0117-0170

AUTHOR INFORMATION

Complete contact information is available at:
<https://pubs.acs.org/10.1021/acsenenergylett.0c01977>

Notes

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