



Toward better batteries: Solid-state battery roadmap 2035+

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ABSTRACT

Recently, *Solid-State Battery Roadmap 2035+* was released by Fraunhofer ISI, which supports the German battery research. As part of the accompanying project BEMA II funded by the Federal Ministry of Education and Research (BMBF), the roadmap comprehensively summarizes the current and future developments of solid-state batteries at the material, component, cell and application level, benchmarking with the anticipated developments of Li-Ion Batteries (LIBs) in the time frame of next ten years. This perspective will briefly summarize the main points of the roadmap and make some corresponding discussions.

1. Introduction

Lithium-ion battery has been the dominating energy storage technology since its first commercialization in 1991, but gradually approaches its energy density limit and demonstrates potential safety risks. Solid-state batteries (SSBs) with solid electrolytes (SEs) to replace organic flammable liquid electrolytes (LEs) can ultimately solve the safety problems and hopefully improve key battery performances [1,2]. In May 2022, Fraunhofer ISI has developed *Solid-State Battery Roadmap 2035+* [3]. Fraunhofer ISI is supporting the German battery research with a roadmap and monitoring process, strategic information processing and status seminars for the exchange of information on scientific progress and technology transfer. More than 100 experts mainly from Germany and Europe contributed to the roadmap. Based on an extensive literature review and an in-depth expert consultation process, the roadmap critically evaluates existing research as well as the latest findings and compares the development potential of solid-state batteries over the next ten years with that of established lithium-ion batteries. From a macro perspective, the most promising SSB development routes are predicted and detailedly discussed. The current and future developments at the material, component, cell and application level are summarized and critically discussed, while benchmarking with the anticipated developments of LIB in a short (<2025), medium (<2030),

and longer (>2030) timeframe.

The roadmap is centered around five themes: 1) introduction of the major components in SSBs (i.e., anodes, cathodes and SEs) and respective interfacial compatibility. 2) production process of SSB components and cells, along with material availability and economic/sustainability aspects. 3) promising SSB cell configurations (Li-oxide, Li-sulfide, Si-sulfide and Li-polymer). 4) international industry perspectives of SSB. 5) the full SSB roadmap with steps from materials to cells and cells to applications. This perspective will briefly summarize these themes and make some corresponding discussions. Besides, some suggestions are given for the future development of solid-state batteries in China.

2. Key materials in SSBs

Solid-state battery mainly consists of a solid electrolyte separator, anode and cathode active materials. The most promising anode active materials to achieve high energy density are lithium metal and silicon. According to the roadmap, lithium metal has the highest technological potential as the anode material, followed by silicon, which has a higher market application potential than lithium counterparts. Although alloy anodes such as Li-In and Li-Al have good compatibility with SEs in laboratory research, they are not discussed in this roadmap. For cathode active materials (CAMs), the roadmap points out that conventional

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layered oxide materials (e.g., lithium nickel manganese cobalt oxides (NMC), lithium nickel cobalt aluminum oxides (NCA), lithium cobalt oxide (LCO) have the highest technological potential for application in SSBs, followed by medium-price and medium-performance materials such as lithium iron phosphate (LFP), or high-voltage materials like Li (Mn, Ni)₂O₄ (LMNO). Due to its low cost and high safety, LFP gains as much recognition as layered oxide materials from a market perspective.

As for the three main groups of solid electrolyte materials, i.e., oxides, sulfides and polymer SEs, their advantages and challenges have been detailly discussed in this roadmap. Fig. 1 is a brief summary of the relevant content. Garnet-type SEs represent the most promising type among oxide SEs due to their wide electrochemical stability window. For sulfide SEs, argyrodites such as Li₆PS₅Cl are most promising owing to their high ionic conductivities and high kinetical stability toward Li metal. Another promising inorganic SE family is metal-halide SEs (Li-M-X, M = metal, X = F, Cl, Br, I), which have gained increasingly more attention in recent years due to their excellent compatibility with high-voltage cathode materials [4] and suitability as catholyte in SSB. Compared with inorganic SEs, flexible polymer SEs have better interfacial contact with electrodes, lower cost and ease of large-scale manufacture. Combining the complementary advantages of polymers and inorganic SEs, composite polymer electrolytes (CPEs) have been widely investigated in recent years. The introduction of inorganic fillers (especially inorganic SEs) in SPEs can greatly improve their RT ionic conductivity. Furthermore, the stability of polymer electrolytes toward Li anode and high-voltage cathode can be improved for extended electrochemical window. Up till now, no single type of SE can satisfy all the strict requirements put forward by high-energy-density SSBs. Solid electrolytes still face many challenges, among which the processability and (electro-)chemical compatibility are the main challenges for oxide and sulfide SEs, respectively. Although polymer SEs are already implemented in some applications, it is difficult to construct high-power and high-energy density SSBs by polymer SEs due to their low ionic conductivities. In addition to the problems of the solid electrolyte itself (e.g., air stability, ionic conductivity, etc.), most of the problems occur at the interface between different components. At the CAM/SE interface, the contact loss during cycling and parasitic interfacial reactions are two main issues. At the anode/SE interface, the continuous growth of SEI seriously deteriorates the electrochemical performance. Moreover, the Li dendrite penetration problem is almost inevitable for most SEs if Li metal anode is applied. Aiming at these problems, many strategies have been proposed by fundamental research. The modifications of SEs like introducing inorganic additives, microstructure design and doping strategy are promising avenues for improving their intrinsic (electro)

chemical stability and ionic conductivity. In-situ or ex-situ formed interlayers between SE and electrodes are always needed to prevent interfacial side reactions and Li dendrites growth. Furthermore, the optimizations of composition, morphology, macro/micro-structure for electrode composites are also very important to maintain intimate ionic contact and improve capacity performance [5]. Recently, designing asymmetric SEs with heterostructures (e.g., anode/sulfide SE/halide SE/cathode) is a promising route due to the Janus property, which can satisfy the different stability requirements from the cathode and the anode, respectively [6].

3. Production process of SSBs

The production processes of SSBs are classified into three steps, i.e., the electrode and electrolyte membrane production, cell assembly and cell finishing. The process chains of electrode/electrolyte membrane consist of components mixing, layer forming and layer compaction [7]. More specifically, the production methods can be divided to two kinds, i.e., solvent-free method (e.g., dry calendering, dry spraying, extrusion and vapor/aerosol deposition) and wet processing (e.g., slurry/tape casting and solution infusion). According to the roadmap, the main processing approaches are summarized in Fig. 2. The compatibility of each method with various SSB components has been labelled out in the figure.

Slurry/tape casting is a widely used and high-throughput process that is compatible with most materials in SSBs. Instead of free-standing SE separators, cathode-supported SEs are more suitable for this manufacturing process, as thinner separators can be acquired and automated manufacturing is highly desirable for large-scale SSB fabrication. However, the limited options of solvents and binders for sulfide SEs and high-temperature sintering process for oxide SEs remain to be the main challenges for this method. In addition, some more disadvantages still exist, including but not limited to the extensive and costly processes of solvent recovery and film drying. Solution infusion is a supplementary approach for wet processing and it has been developed recently for thin sulfide/hybrid SE film preparation. This method has only been used in laboratory cells so far and its scalability remains unknown. Solvent-free methods can prevent drying/solvent recovery process, and have unique compatibilities with all-solid-state lithium batteries (ASSLBs) as they allow SEs (especially sulfide SEs) to maintain high ionic conductivities by avoiding detrimental organic solvents. Among various dry methods, dry calendering and dry spraying are the two most promising technologies that can realize large-scale mass-production of batteries, because they are suitable for roll-to-roll production.

Oxide Electrolytes	Sulfide Electrolytes	Polymer Electrolytes
Garnet ☆☆☆	Argyrodites ☆☆☆	Polymer Matrix
NASICON ☆☆☆	LGPS sub-class ☆☆☆	+ Lithium Salt
LiPON ☆☆☆	LPS sub-class ☆☆☆	+ Additives
Perovskite ☆☆☆	Thio-LISICONs ☆☆☆	
✓ Good mechanical stability ✓ Good chemical stability ✓ Insensitivity to temperature ✗ High sintering temperature ✗ Brittleness ✗ Low Li metal stability ✗ Low ionic conductivity as a catholyte ✗ Thin film processing	✓ The highest conductivity ✓ Softness and plasticity ✗ Instability toward lithium metal ✗ Instability toward high-voltage cathode ✗ Manufacturing of SE ✗ Precursor/material availability (e.g. Li ₂ S) ✗ Instability in air	✓ Low cost ✓ Easy processing ✓ High flexibility ✗ Resistance to dendrite formation ✗ Limiting current density ✗ Low ionic conductivity ✗ Instability toward high-voltage cathode

Fig. 1. A brief summary of solid electrolyte materials discussed in the roadmap. The colors of the arrows represent the severity of the challenge. The redder the color, the higher the severity. (For interpretation of the references to color in this figure legend, the reader is referred to the Web version of this article.)

	Solvent-free				Wet processing	
	Dry calendering	Dry spraying	Extrusion	Vapor/aerosol deposition	Slurry/tape casting	Solution infusion
Cathode Composite	✓	✓	✓	✓	✓	✗
Li metal	✗	✗	✓	✓	✗	✗
Si/C	✓	✓	✓	✓	✓	✗
Oxide SE	✗	✗	✗	✓	✓	✗
Polymer/Hybrid SE	✓	✓	✓	✗	✓	✓
Sulfide/Halide SE	✓	✓	✗	✗	✓	✓

Fig. 2. Main processing approaches for cathodes, anodes and SE membranes in SSBs [7–9].

However, few papers/patents have been published/filed in the field of SSBs so far [8], and the film uniformity and scalability remain challenging. New sophisticated facilities for scale-up manufacturing need to be developed as well as better binders. Extrusion process is already established and used commercially for polymer electrolytes and thin lithium foils. However, for inorganic SEs, some technical barriers still remain owing to their hardness and lower malleability. Vapor/aerosol deposition is an alternative method for electrode preparation and it is especially suitable to acquire high quality and dense oxide SE layers without high-temperature sintering steps. It is worth to note that this method is still in its infancy, mass-production is not applicable due to the high cost and complex operations.

The most suitable cell format of currently produced SSBs is in planar shape (prismatic or pouch cells), which can retain the structural integrity of SEs. Full SSB cells can be assembled by stacking the anode, solid electrolyte, and cathode on top of each other. For multilayer cells, bipolar configuration can be adopted to achieve smaller pack resistance and higher energy density. Compared to liquid-electrolyte LIB, electrolyte filling and degassing steps can be avoided so that the SSB formation and aging time can be shortened. Moreover, considering the contact losses or dendrites formation during cycling of inorganic SE-based SSB, a stack pressure of at least several MPa is required, which may need additional devices.

4. Promising SSB roadmap

At the full cell level, there are five key performance indicators (KPI), which are safety, energy density, fast charging ability, long-term stability/lifetime, and price. SSBs have higher safety than liquid LIBs, while the safety risk cannot be completely avoided if short circuits occur. Research on thermal stability of SSBs is still in its initial stage now [10]. Due to the immobility of the anionic framework in solid lithium superionic conductors, there is no bulk or concentration polarization, leading to enhanced kinetics and higher power capabilities [11]. Since fast charging relies on high ionic conductivity and sufficient interfacial contact, sulfide SEs have a higher potential for fast charging than oxide SEs. The lifetimes of SSBs are mainly determined by the electrode/SE interfacial stability. The volume of most electrode materials changes greatly during cycling, contributing to void formation at the interface and mechanical degradation. Therefore, flexible polymer-based SSBs may enable longer lifetimes. Li dendrite growth is another factor affecting long-term stability of SSBs. As for the price of SSB, material costs account for the largest share. The cost of some inorganic SEs can be estimated by their elemental cost, such that SEs (e.g., $\text{Li}_{1-x}\text{Al}_x\text{Ge}_{2-x}(\text{PO}_4)_3$ and $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$) containing precious metals are costly. Considering the immature supply chains, the precursor materials

may be much more expensive than their elemental cost, such that it is difficult to precisely estimate the final material costs. The cell manufacturing costs also account for a large part, and the manufacturing facilities for liquid LIBs are not fully compatible with SSB production. For example, the inert atmosphere and the sintering process required for sulfide and oxide SEs all lead to additional costs. Polymer SSBs can be cost-effective and economically competitive if production lines are established and scaled up.

A large number of SSB cell concepts have been investigated in the laboratory, among which several promising concepts have been proposed according to the roadmap. Based on oxide SE, Li/Oxide SE/NMC + Gel is a promising concept/configuration. Considering the low conductivity and limited interfacial contact, it is challenging to use oxide SEs as catholytes, thus gel/liquid electrolytes in combination with oxides are often used as catholytes. At present, some Chinese companies like WeLion, ProLogium and QingTao have adopted this hybrid concept and its validity has been verified. In long-term research, using high ion-conductivity sulfide SEs or halide SEs as catholytes is potentially possible to construct ASSLBs, while there are few related studies at present [12]. As for anode, the application of silicon-based anode materials is relatively mature. Li metal anode also has very high feasibility and QuantumScape has already developed anode-free Li metal technology. However, considering the limited production methods for pure oxide SE films, large-scale production and commercialization are at risk. As shown in Fig. 3, Li/Oxide/Gel@NMC811 cell concept is expected to reach energy densities of 315 Wh/kg and 1020 Wh/L in the medium term (2025–2030). At around 2035, with the application of

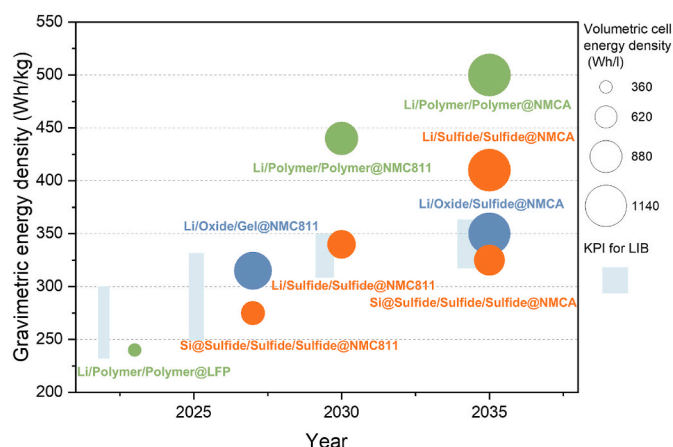


Fig. 3. Promising SSB cell concepts and roadmap.

high-capacity NMCA-type ($\text{LiNi}_{1-x-y-z}\text{Co}_x\text{Mn}_y\text{D}_z\text{O}_2$ with $x < 0.05$, $(1-x-y-z) > 0.9$ and $D = \text{Al or Mg}$) CAMs, Li/Oxide/Sulfide@NMCA cell concept is expected to reach energy densities of 350 Wh/kg and 1140 Wh/L.

Based on sulfide SEs, Si/Sulfide/Sulfide@NMC and Li/Sulfide/Sulfide@NMC are two promising concepts, which may come to market in medium term. Compared with oxide SEs and polymer SEs, sulfide SEs with high ionic conductivity and ductility are more likely to achieve high-energy-density ASSLBs. At the cathode side, owing to the low oxidation potentials of sulfide SEs, the CAM particles are always coated with ionically-conductive buffer layers. Another promising method that should be supplemented is using halide SEs as catholytes, which can prevent the introduction of coating materials and improve capacity performance [13]. For silicon-based anodes (e.g., silicon/carbon composite and pure silicon), their production methods and fast charging capability are closer to market maturity than Li metal anode. The biggest challenge is the huge volume change of silicon during cycling, which is being solved through structural design and binder optimization [14]. According to the prediction, Sulfide@ Si/Sulfide/Sulfide@NMC811 cell concept is expected to reach energy densities of 275 Wh/kg and 650 Wh/L in the medium term (2025-2030) and Sulfide @Si/Sulfide/Sulfide@NMCA cell concept is expected to reach energy densities of 325 Wh/kg and 835 Wh/L at ~2035. Replacing silicon-based anode with a Li metal, Li/Sulfide/Sulfide@NMC811 cell concept is expected to reach energy densities of 340 Wh/kg and 770 Wh/L at ~2030, and Li/Sulfide/Sulfide@NMCA cell concept is expected to reach energy densities of 410 Wh/kg and 1150 Wh/L at ~2035. Although some sulfide SEs like argyrodites are kinetically stable with Li metal, Li dendrite problem is almost inevitable. From this aspect, elemental doping and introduction of an interfacial layer have become the main solutions.

The polymer SSB cell concept consisting of Li metal anode and LFP cathode has already been on the market for several years. Compared with SSB with inorganic SEs, polymer SSB maintains a lower price. However, it can not compete with state-of-the-art liquid LIBs due to its moderate energy densities (240 Wh/kg and 360 Wh/L) and high operation temperature (50-80 °C). To further improve the energy density of polymer SSBs, the polymer SEs should be optimized to improve the room-temperature ionic conductivity and electrochemical stability toward high-voltage CAMs. Constructing passivating layer at cathode/electrolyte interface or designing inorganic-polymer composite SEs [15] are possible solutions. Li/Polymer/Polymer@NMC811 cell concept is expected to reach energy densities of 440 Wh/kg and 900 Wh/L at ~2030 and Li/Polymer/Polymer@NMCA cell concept is expected to reach energy densities of 500 Wh/kg and 1150 Wh/L at ~2035.

Although liquid LIBs currently exhibit higher energy densities (230-300 Wh/kg and 600-750 Wh/L) than SSBs, the energy densities of SSBs might potentially be two times higher than today's liquid LIBs in the next decade by applying Li anodes and high-capacity CAMs. In terms of the market size, the overall liquid LIB market is expected to grow from 400 GWh to 0.5-2 TWh by ~2025, 1-6 TWh by 2030 and 2-8 TWh by ~2035. The annual production capacity for polymer SSB is expected to increase from 2 GWh to 10-50 GWh by 2035, 0 to 20-50 GWh for sulfide SSB, and 0 to 10-20 GWh for oxide SSB. It is indicated that liquid LIB will still dominate the market for a long time, and SSB is expected to appear as a supplementary application in some areas due to its initially higher costs. For example, polymer SSBs are suitable for electric buses, automatically-guided vehicles and passenger cars/trucks in the future. Sulfide and oxide SSBs will be used in passenger cars first and then in trucks or electric passenger aviation.

In sum, the roadmap proposes ambitious goals/estimations (especially the energy density) to help assess the technological potentials of SSBs, although these estimations are still highly speculative. Due to the strategic need for alternative battery technologies, SSBs will obtain continuously increasing attention. At present, during transition from liquid LIB to ASSLB, quasi-solid-state batteries with a small amount of liquid electrolyte are already on the eve of mass production. Moreover,

as disclosed by some companies like Welion, QingTao and ProLogium [16], the energy densities of their quasi-solid-state batteries all exceed 300 Wh/kg and are comparable to the state-of-the-art liquid LIBs.

This roadmap shows the optimism and ambition of Germany and Europe Union towards the future of SSBs. It is predictable that German government and joint European projects will invest more in the R&D of SSBs. China has ranked first in the number of peer-reviewed publications, but Chinese companies still need to catch up with and learn from pioneering companies like Samsung SDI and Toyota, etc. Up till now, it is still controversial which technical route will ultimately provide the best-performance solid-state battery, which means that different approaches have to be developed in parallel. Therefore, significant private/public funding/investment that goes beyond mere research funding is needed to minimize the gap between Chinese and Japanese, Korean or U.S. players in production technologies, pilot production, start-up and industrial activities. Fortunately, from the information disclosed, China seems to have taken a leading position in the industrialization process in terms of quasi-solid-state batteries around the world. Based on existing industrial advantages in China, it is advisable to promote production-study-research combination, and form a good development ecology of government guiding, scientific research institutes powering, capital following up, and enterprise landing.

5. Conclusion

The merits of solid-state batteries are widely discussed in recent years, and related research has also grown explosively. However, commercial SSB for high-volume auto-motive applications is not available and many problems have yet to be studied and solved. Fortunately, based on recent progress in scientific research and industrialization exploration, the feasible roads to final success are turning more obvious. The roadmap demonstrates that solid-state batteries have a lot of potential, but will have to prove their commercial viability in the next five years. Unified planning, continuous funding from official organizations and cross-sectorial research efforts across the battery community are highly recommended to promote the more comprehensive and rapid development of SSBs.

CRedit authorship contribution statement

Dengxu Wu: Visualization, Methodology, Formal analysis, Writing – original draft. **Fan Wu:** Conceptualization, Validation, Writing – review & editing, Project administration, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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