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A Roadmap for Solid-State Batteries

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Solid-state batteries are considered as a reasonable further development of lithium-ion batteries with liquid electrolytes. While expectations are high, there are still open questions concerning the choice of materials, and the resulting concepts for components and full cells. On the basis of an analysis of all materials and concept options, a roadmap for solid-state batteries is presented, relying on both literature survey and experts' opinions. Diverse cell concepts with different solid electrolytes may be developed up to the commercial level, yet there are still major uncertainties concerning production routes, safety as well as cost. As one of the key developments, it appears that hybrid material and cell concepts may be particularly successful on the way to commercialization.

1. Introduction

Electrification of transportation is considered as one key ingredient on the way to reduce CO_2 emission (as well as other emissions) and environmental impact, thus to fight climate change and other environmental issues.[2] In parallel, electrochemical storage is as well considered as an important technology to stabilize the future electric grid with its much higher fraction of electric energy supplied from renewable sources. These developments lead to fast growing markets and high investments in battery R&D and continuous performance increases, that is, increases in specific energy, power, cycle lifetime as well as reduction of costs. Despite these achievements, current battery technologies still have their limits, as there is a trend to offer wide-range (>500 km) electric vehicles (EV). This can only be solved by larger (heavier) battery packs and/or batteries with higher energy density. Volume and weight can be saved by more efficient engineering of the battery pack, for example, through so-called cell-to-pack concepts.[3] Higher energy density at cell level is another way to increase battery capacity or reduce footprint. Unfortunately, higher energy densities also increase safety risks to the flammable liquid electrolyte (LE) in state-of-the-art lithium-ion batteries (LIBs) and pose a higher risk of fire in case of accidents. As a consequence, R&D efforts in next-generation battery technologies consider solid-state battery (SSB) cell concepts as one of the most promising alternatives to state-of-the-art LE LIB, promising higher energy densities and higher safety at the same time (**Figure 1**).

The last decade has seen fast accelerating research on the material and component level for SSB.[4] Key limitations of LIB that are addressed in the development of SSB comprise a) energy density and specific energy, b) power density, c) safety, and d) temperature window during operation. Overcoming these limitations is approached by extensive investigations on a) the use of lithium (and silicon) anode, b) the use of single-ion solid electrolytes (SE) that avoid concentration polarization, c) reducing the weight fraction of flammable LE, and d) the use of thermally stable SE. Clearly, the development of high-performance LIB is continued, and thus, they are "moving targets" for the development of SSB.

Different classes of SE have been investigated, and new or improved electrolytes are still being developed, as their requirements are manifold. While particularly the ionic conductivity was considered as primary key performance indicator (KPI) at the beginning, now other KPI like stability in contact with anode and cathode materials, mechanical stability, thermal stability, to name a few, are considered of importance, too. Cost of SE synthesis will surely be an important issue and may become a critical factor, as inorganic SE have at least a factor of 10 higher lithium concentration than organic electrolytes (either liquid or polymer).[4] Conventional SSB cell concepts rely on a single SE,[5] which may not be the ultimate and successful choice.[4] As there is not the one-and-only SE with equally high values for all KPI, hybrid materials or cell concepts are being developed and may be successful. Due to these uncertainties concerning the most suitable material combinations, processing and production steps may also vary depending on the materials choice.

In this work, we like to have a look at the bigger picture, considering the future of SSB from the materials level via the

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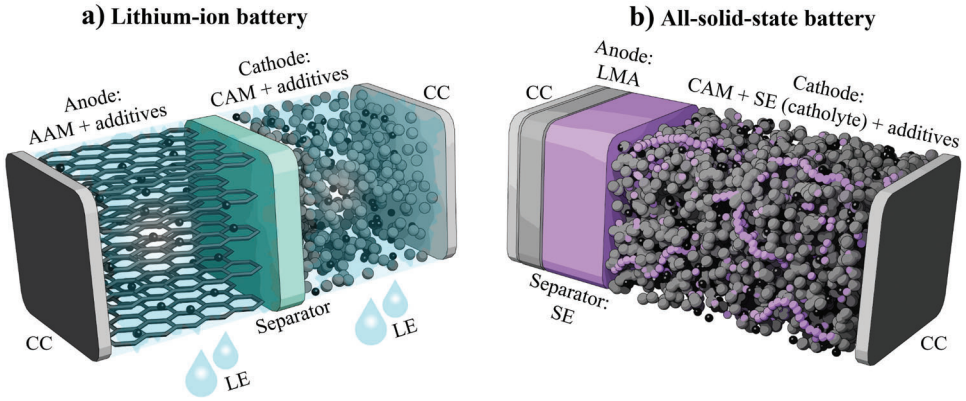


Figure 1. Graphic illustrations of a) a state-of-the-art lithium-ion battery with liquid electrolyte and b) an all-solid–state battery with lithium metal anode. (CC: current collector; LE: liquid electrolyte, SE: solid electrolyte; AAM/CAM: anode and cathode active material, respectively; LMA: lithium metal anode).

component and cell level to production and applications. With this we target for an objective and balanced view into the complex landscape of SSB development and try to help the reader to better understand the opportunities and challenges. It will become apparent that the SSB will most probably not cause a radical change but may rather offer a continuous evolution of LIB, aiming for improved performance and safety. Whether this will be possible also at acceptable cost, is an open question. In general, we will also highlight that the process of technology roll-out in the automotive industry with long periods of testing, qualification, and product design will strongly influence the parallel improvement of LIB and SSB concepts. This perspective is based in parts on our previously communicated report *Solid-State Battery Roadmap 2035+*,[1] but is more concise to reach a broader audience, more aiming at the research community and catches up on new or accelerating developments of the last year, e.g., the trend of hybrid liquid/solid and hybrid solid/solid electrolyte use in batteries.

The results are based on literature data, and expert information that were compiled in the course of a publicly funded R&D project via interviews, an online survey, and an expert workshop.[1] Involved experts came from both academia and industry (cell manufacturer, battery industry, OEM), with a focus on experts from Germany and Europe. In section 2, we will describe all components of SSB, with a special focus on SE. In section 3, we will then consider the most relevant cell concepts, highlighting the advancement of hybrid cell concepts. Production issues are summarized in section 4, and the resulting roadmap is presented in section 5. All relevant numbers and methods can be found in detail in the Supporting Information.

2. Components

2.1. Anodes

Graphite intercalation compounds are by far the most common anode active materials (AAM) in LIB, today, due to a high practical specific capacity (≈360 mAh g^{-1}), low operating potential $E_H \approx 0.1$ V versus Li^+/Li, high availability, safe operation, and rather low cost.[6–8] The function of graphite-based AAM was demonstrated using different concepts for SSBs during the last two decades.[9–11] However, specific energies of SSB higher than those of LIB can only be achieved, if anodes with higher capacity and low potential are implemented. Fast charging with minor capacity fading requires reversible high-rate operation of the anode, which is limited by the graphite anode in LIB.[12] Driven by these requirements and described by the corresponding KPI, which are particularly important in the EV sector, the demand for high-performance AAM increased massively. Lithium metal and silicon-based AAM (e.g., SiO_x or silicon micro- or nanoparticles) are the most obvious and promising choices for SSB concepts, due to their high theoretical specific capacities ($q_{th}(Li) = 3862$ mAh g^{-1}, $q_{th}(Si) = 3579$ mAh g^{-1}) and low operating potentials ($E_H(Li^+/Li) = -3.04$ V vs standard hydrogen electrode, $E_H(Si/Li_xSi) = 0.2–0.4$ V vs Li^+/Li).[13–16] In fact, implementation of the lithium metal anode (LMA) with the described benefits is one of the major driving forces behind SSB research, since a reliable and safe operation might be achieved more conveniently and reliably than with LE.[17,18] Lithium reservoir-free cell concepts, where lithium is directly plated onto an anode substrate (i.e., current collector, CC) during charge, received attention because material and manufacturing costs could be reduced and energy density improved. Every battery cell inherently contains both, an anode and a cathode CC. In cells with a lithiated cathode material, a reversible LMA can be formed during the initial charging step at the anode CC. Once no lithium metal is added before cell assembly, such cells are often denoted as "anode-free" or "anode-less." As any electrochemical cell always has two electrodes, these terms are electrochemical misconceptions, and we prefer to call such cells more precisely as "lithium reservoir-free cells."

For this approach, however, specifically designed CC are required, since lithium deposition is strongly influenced by the substrate surface morphology and its chemical properties.[19–22] A major challenge of lithium metal and silicon-based high-energy anodes is the massive volume change they experience during cycling, thus requiring further concepts for the AAM and SE separator interface to counteract performance fading by mechanical stress.[17,23] Although unwanted lithium dendrite growth can be mitigated to some extent by replacing the LE with SE, LMA suffer from inhomogeneous lithium plating and stripping leading to interfacial contact issues at and pore formation in the SE.[24,25] However, the resistivity of an SE separator against lithium

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dendrite nucleation and growth results from the complex interplay of various parameters, and the root causes are not fully understood, yet. Factors affecting dendrite resistivity are i) the properties of the SE separator itself, such as its mechanical strength (e.g., fracture toughness, Young's and shear modulus, and yield strength), pore density and distribution, homogeneity of grain boundary contacts in the SE, and chemical stability; ii) the adhesion properties at and characteristics of the interfaces between Li metal, SE separator, and the solid electrolyte interphase (SEI), such as the homogeneity of the interfaces, defects at the interfaces, and local electrochemical stability; and iii) the electronic properties of SE grain boundaries and dislocations.[4,26,27] For example, non-uniform charge distribution and local charge transfer resistance facilitate local current constrictions and/or confinements, which propels dendrite formation.

Promising concepts involve application of composites such as LMA with carbonaceous compounds to keep the anode in intimate contact with the SE during cycling without applying excessive stack pressure—at the cost of lower specific capacity.[28,29] Considering high-power anodes, $Li_4Ti_5O_{12}$ (LTO) is a promising option offering long-term stability as volume changes are <1% during cycling. However, a relatively low specific capacity of q_{th}(LTO) = 180 mAh g^{-1} and rather high operating potential of $E_H(Li_4Ti_5O_{12}/Li_7Ti_5O_{12})$ = 1.55 V versus Li^+/Li result in a low energy density on cell level making LTO a candidate for high-duty SSB rather than for the wider mass markets.[30,31]

Graphite anode concepts are actively pursued in SSB R&D, although the gain in energy density is rather small. Anyhow, certain cell concepts allow fast charging of such SSB cells, for example, by employing SE with very high effective ionic conductivities.[32] However, to eventually overcome the physicochemical limits of conventional LIB in terms of energy density and specific energy, stable and safe operation of LMA or Si anodes is paramount and necessitates solidification of the electrolyte at least at the anode side.

2.2. Cathodes

Essentially the same cathode active materials (CAM) used in LIB with LE are conceivable for or are already implemented in lithium-based SSB. Hence, similar technological, ecological, and economic aspects drive CAM research for SSB. Key parameters are cost, raw material availability and criticality, high capacity, long cycle-life, fast-charging compatibility, and high-voltage stability. The specific energy of SSB cells is generally very much controlled by the CAM since relatively low specific capacities are achievable compared to the anode side, resulting in the need for thick cathodes.[33,34] Depending on chemical and processing compatibilities, additional technical prerequisites need to be considered in SSB. The design of CAM and catholyte is paramount if the LE is replaced by an SE, to tackle challenges related to (chemo-)mechanical, (electro-)chemical, and (micro-)structural interplay of CAM with SE, such as charge transport (e.g., electronic and ionic percolation) or interfacial stability issues.[33,35] Based on application and cost-sector, relevant CAM can be divided into three classes. 1) High-energy CAM, in particular nickel-rich layered oxides ($LiNi_{1-x-y}Mn_xCo_yO_2$, NMC; $LiNi_{1-x-y}Co_xAl_yO_2$, NCA; $LiNi_{1-x-y-z}Mn_xCo_yAl_zO_2$, NMCA) with mean operating potentials $E_H \approx 3.8$ V versus Li^+/Li and high theoretical capacities up to $q_{th} \approx 275$ mAh g^{-1} ($LiNiO_2$, LNO), trigger relentless research for (premium) EV with enhanced driving range.[36–39] However, structural instability at higher degree of delithiation limit their reversible capacity for practical applications.[40–42] Thus, (electro-)chemical stabilization strategies such as doping, coating, or gradient compositions are necessary.[42] One established benchmark is $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ ($q_{pr} \approx$ 170–180 mAh g^{-1}), but a higher nickel fraction is aimed for and, for example, $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ ($q_{pr} >$ 200 mAh g^{-1}) and $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ ($q_{pr} \approx$ 200 mAh g^{-1}) have high technological importance with more potential to come, once suitable protective coatings or stabilizing dopants are applied to improve interfacial stability between CAM and SE.[37,43,44] Even higher energy densities can be achieved with layered lithium-rich oxides (LLO), but severe capacity fading still hinders practical application.[42,45] 2) High-voltage CAM, for example, spinel-type oxides $LiMn_{2-x}Ni_xO_4$, offer operation at high potentials (up to $E_H \approx 4.6$ V vs Li^+/Li) with good cycling stability and rate capability, but seem less relevant for the automotive mass market due to their limited capacity (e.g., the theoretical capacity of LMO is q_{th} = 148 mAh g^{-1}).[46–49] 3) Lower cost CAM, especially olivine-type $LiFePO_4$ (LFP) and $LiMn_xFe_{1-x}PO_4$, retrieved vast attention from automotive OEM.[50,51] Although their energy density is limited by a practical capacity $q_{pr} \approx$ 160 mAh g^{-1} (LFP) and an operating potential $E_H \approx 3.3$ V versus Li^+/Li, the cost-factor, safe operation, thermal stability, that is, requiring less cooling at pack level, and high cycling stability revived LFP application for lower cost LIB in EV and is also discussed for SSB.[52–55] LFP is particularly interesting in combination with certain polymer SE, which are oxidized in the potential window of NMC and NCA cathodes, such as poly(ethylene oxide) (PEO).[56–58] Besides, conversion-type CAM are researched, for example, iron and copper sulfides or elemental sulfur, and promise to combine low cost and high energy density. Yet, their technological readiness level remains low due to unsolved failure mechanisms mainly driven by kinetic issues, side reactions and extreme mechanical stress during de-/lithiation caused by large volume changes.[59,60]

Due to anticipated higher costs of SSB compared to existing LIB, we expect SSB to enter the market as premium cells for premium applications, making the cost factor of cheaper CAM with small specific capacities such as LFP less relevant in the short term for SSB technologies. From technological and scientific point of view, NMC and NCA with high Ni content are currently the most viable CAM for competitive high-energy SSB cells to outperform state-of-the-art LIB and we expect them to dominate future development of SSB cells. Practical application of LLO, which allow even higher energy densities, could be an option if their issues of fast cell fading can be solved.

2.3. Solid Electrolytes

Three main groups of SE materials receiving most attention in recent SSB research are polymer, sulfide, and oxide SE. It should be mentioned that we use the terms "sulfide SE" and "oxide SE" for the sake of readability even though they do not represent the correct terminology for many materials in these classes (e.g., for lithium thiophosphates). Sulfide and oxide SE can be used in

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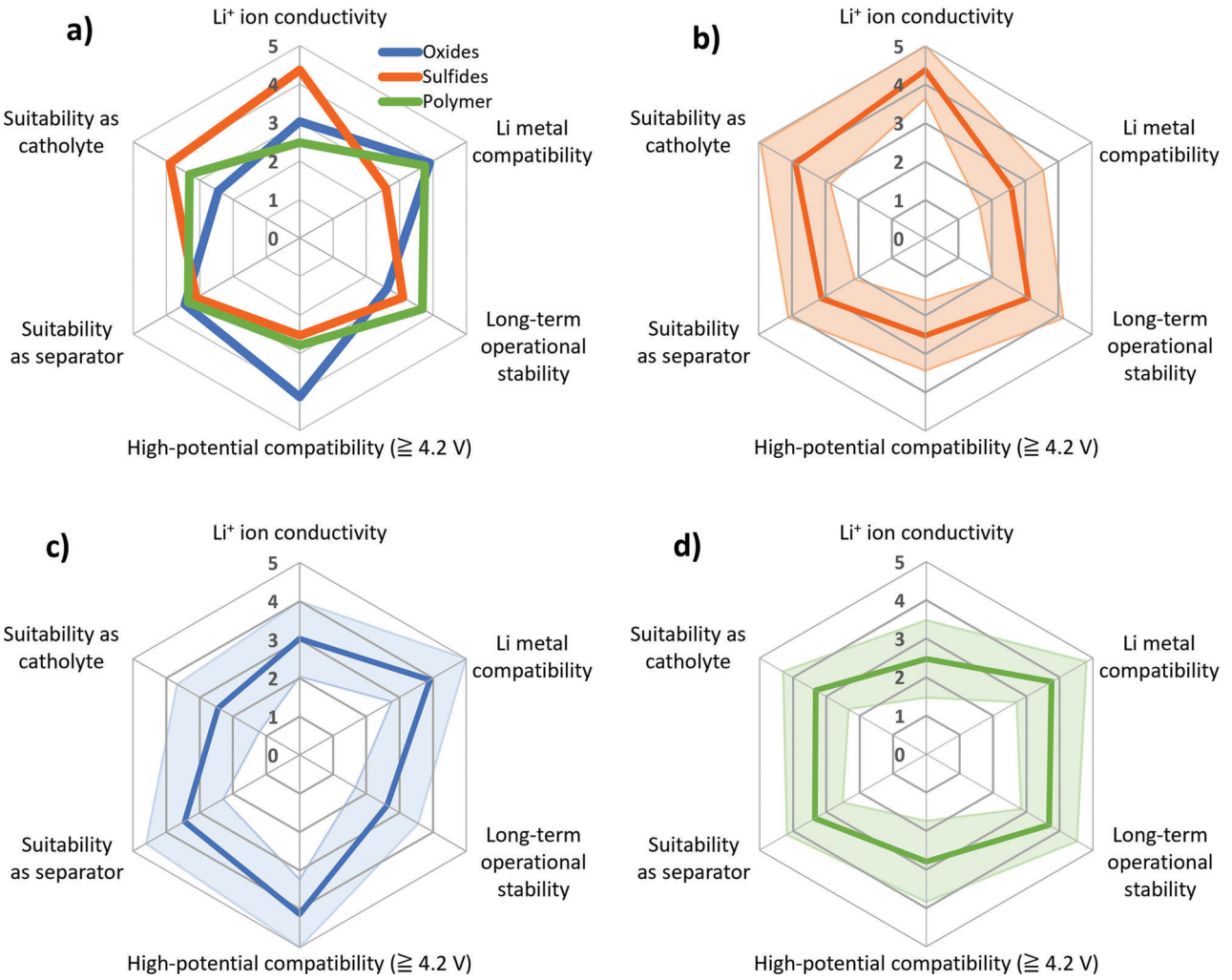


Figure 2. Radar charts of properties a) for the three main classes in comparison, b) for sulfide SE, c) for oxide SE, and d) for polymer SE. The values (1: poor, 2: rather poor, 3: medium, 4: rather good, 5: good) represent an expert assessment (details are provided in the Supporting Information).

glass, crystalline, or glass-ceramic states, resulting in manifold properties. They cover several types of compounds, all containing lithium and either sulfur or oxygen in addition to further elements such as phosphor, silicon, germanium, or halides (sulfide SE) and phosphor, titanium, aluminum, lanthanum, germanium, zinc, or zirconium (oxide SE). In contrast, polymer SE are composed of either polymer/lithium salt complexes or single-ion–conducting polymers.[61] At room temperature (RT) they are semi-crystalline or fully amorphous,[62,63] but operating temperature is usually above glass transition or melting temperatures and thus they have similarities to LE.

An ideal SE candidate should exhibit a high Li^+ ion conductivity, wide electrochemical stability window (ESW) without redox decomposition at either the anode or cathode, sufficiently high elastic modulus and yield strength to reduce cracking, high resistance against lithium dendrite formation and penetration, low interfacial resistance, and good adhesion with both electrodes.[64] As essential indicators for these properties, the i) lithium-ion conductivity at RT, ii) lithium metal compatibility, iii) long-term operational stability, iv) high potential compatibility, v) suitability as separator, and vi) suitability as catholyte of the three main SE classes are compared in **Figure 2**.

Previous simulations demonstrated that an effective lithium-ion conductivity of $\sigma \approx 10$ mS cm^{-1} should be targeted for the solid catholyte to achieve reasonable energy densities at enhanced C-rates, for example, cycling at 1C with 100 µm cathode thickness.[65] In addition, high-energy ASSB cells require internal resistances $\lesssim$40 Ω cm^2 and separator thicknesses $\lesssim$50 µm to allow cycling at 1C, hence a lithium-ion conductivity $\approx$0.1 mS cm^{-1} is aimed for the SE separator.[34] These quantitative limits for cycling at 1C and net ionic conductivities of promising examples are shown in **Figure 3** (cf. Table S1, Supporting Information).

2.4. Sulfide Solid Electrolytes

Many sulfide SE offer ionic conductivities (Figure 3) similar or even better compared to LE, a prerequisite for power and

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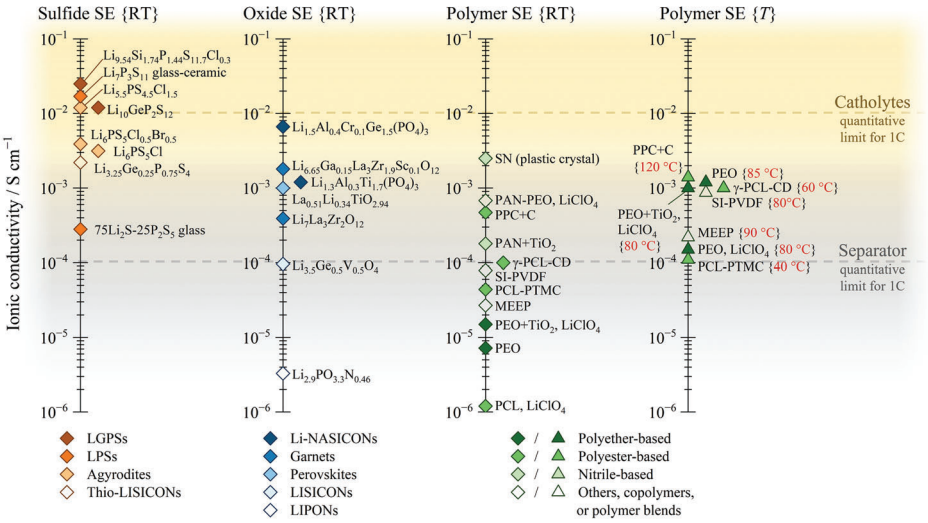


Figure 3. Selected examples of promising SE (sub-)classes and their net ionic conductivities. RT values are given for sulfide,[32,66–72] oxide,[73–79] and polymer SE[80–90] and additionally at elevated temperatures (red) for polymer SE. Various polymer SE combinations were reported in literature, for example, with different lithium salts (LiX), using additives, blending polymers, by copolymerization, etc. and only selected promising examples are given (see main text). Unless otherwise labeled, ionic conductivities are given for LiTFSI polymer complexes. For an extensive picture we refer to reviews on this topic.[91–103] CD = Cyclodextrin, other abbreviations are explained in the main text. Values for typical industrial LE are in the range of 3×10^{-3} to 8×10^{-3} S cm^{-1}.[104,105] All values are summarized in the Supporting Information (Table S1, Supporting Information).

fast-charging capability.[32,65,69,71] Due to that, sulfide SE are currently the most convenient catholyte choice (Figure 2b; cf. quantitative limit in Figure 3). The softness of sulfides allows good interfacial contact to the active materials and, to some extent, compensation for cracking and pore formation during cycling. Also, mechanical processing steps are simplified, even allowing cold pressing and high-pressure calendering. Thus, low grain boundary resistance and good interfacial contacts are achievable, helping to prevent lithium dendrite formation.[106] However, practical application in mass SSB markets is currently limited, because sulfides typically suffer from interfacial instability at the LMA and CAM as well as a rather narrow ESW (Figure 2b).[107,108] Tailoring coatings, doping, and generating artificial SEIs are concepts to protect sulfides from (electro-)chemical decomposition.[109–111] In addition, the particle size of SE is critical and the optimal particle size distribution (PSD) has to be adjusted to the PSD of the active materials. Moreover, large-scale preparation and suitable handling of several sulfides are challenging due to moisture and air instability, potentially causing release of toxic H_2S and SO_2 gases.[112] Lithium-based argyrodites Li_6PS_5X (X = Cl, Br, I) and related SE (e.g., $Li_{5.5}PS_{4.5}Cl_{1.5}$) appear particularly promising due to their high ionic conductivity, conceivable cost-efficient production, and potential compatibility with LMA and most CAM once suitable protection strategies are applied.[67,113–118] Glasses derived from the xLi_2S-$(100-x)P_2S_5$ system (LPS) outperform crystalline sulfides in terms of the stability toward active materials and the ESW, but rather poor ionic conductivities prevent practical application so far.[69,119–121] Glass-ceramic LPS (e.g., $Li_7P_3S_{11}$) offer better Li^+ ion mobility, but have severe stability issues at the cathode and with LMA.[69,70,122–124] $Li_xM_{1-y}M'_yS_4$ (M = Si or Ge; M´ = P, Al, Zn, Ga, or Sb) crystallizing in the γ-Li_3PO_4 structure (Thio-LISICON) have no significant advantage over other sulfides, thus, lower technological relevance so far.[72,125–127] $Li_{10}GeP_2S_{12}$ (LGPS) and related SE (e.g., $Li_{9.54}Si_{1.74}P_{1.44}S_{11.7}Cl_{0.3}$) exhibit the highest ionic conductivities discovered for sulfides until now.[32,71,117] Application of germanium-free (Ge is too expensive for practical use) LGPS-related SE as catholyte seems possible if strategies against the narrow ESW are pursued.

2.5. Oxide Solid Electrolytes

Most oxides are mechanically stable (rigid), insensitive to elevated temperatures, and exhibit wide ESW offering high-voltage applications and limiting decomposition reactions during cycling (Figure 2c).[128–130] Detrimentally, oxides are stiff and brittle, thus requiring massive stack pressure or advanced strategies to ensure sufficient interfacial contact.[131] Although most oxides are chemically stable toward air and moisture,[132] which facilitates processing conditions, bending and rolling processes are inconvenient due to the mechanical properties. To obtain reasonable effective ionic conductivities, cost-inefficient sintering steps are often required for oxide SE, causing also a challenge for co-processing with high-temperature instable CAM.[133] Because a few promising oxides are (electro-)chemically stable toward lithium metal and mitigate dendrite formation, but exhibit too low RT ionic conductivities, they are considered as SE separator rather than SE catholyte (Figure 2c, cf. quantitative limits for catholyte and separator in Figure 3).

Garnet-type oxides seem most promising amongst this group with highest potential for application in SSB and comprise materials like $Li_7La_3Zr_2O_{12}$ (LLZO) and metal-doped LLZO (e.g., Ga, Ta, Al, and Sc).[73,74,134] Good (electro-)chemical stability of most garnet-type oxides with elemental lithium make them compatible separators for high-energy SSB based on LMA concepts.[135] NASICON-type oxides such as $Li_{1+x}A_xTi_{2-x}(PO_4)_3$ (LATP, A = Al

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or various metals) and $Li_{1+x}Al_xGe_{2-x}(PO_4)_3$ (LAGP), show beneficial properties for applications in SSB as well, but are unstable toward lithium metal.[75,76,136,137] $Li_{2+2x}Zn_{1-x}GeO_4$ (LISICON)-type,[138] $Li_xPO_yN_z$,[77,139] and perovskite-type SE, for example, $Li_{3x}La_{2/3-x}TiO_3$,[79,140] have lower market potential because no clear advantages over the other two oxide subclasses are given. Standalone oxide SE are unlikely to come into application in SSB mass markets, but could in niche markets, where mechanically stable and heat-resistant batteries are required, or in combination with other SE or LE (hybrid concepts, see section 3.4).

2.6. Polymer Solid Electrolytes

The Li^+ ion transport in polymer SE occurs through the motion of polymer segments, and thus, strongly depends on the operating temperature and the polymer's glass transition or melting point.[92,141] Polymer SE are typically soft, highly flexible, offer better processability, improved adhesion to electrodes, and enhanced compensation for volume changes compared to inorganic SE.[94,142,143] Also, fabrication is already established on larger scale, potentially cost-efficient, and critical raw materials are usually dispensable. However, challenging roadblocks hamper mass market application (Figure 2d): Many unmodified polymer SE suffer from insufficient (long-term) resistance to lithium dendrite penetration and only provide mediocre RT ionic conductivity (Figure 3), making external battery heating essential.[103,144] Depending on the final application, improving the fast-charging capability (SSB with polymer SE typically require long charging time, see section 3.3), thermal stability, and ESW (limited to <4 V vs Li^+/Li for most polymer SE) could become important. Balancing the trade-offs between mechanical strength/plasticity and ionic conductivity is key to tailor performance-optimized polymer SE. Appealing strategies widely pursued for functional adjustment are i) blending polymer hosts with other polymers, ii) designing block or grafted copolymers, and iii) using additives.[88,91,97,145,146] For example, passive inorganic fillers (e.g., Al_2O_3, SiO_2, TiO_2) are frequently employed to modify polymer properties such as the plasticity and thus, for example, adjust the ionic mobility.[147–150] Also, introducing certain quantities of solids actively taking part in the ionic conduction mechanism or liquid components are popular approaches (see hybrid concepts, section 3.4).

As polymer scaffold, PEO is widely used, mainly due to a low glass transition temperature, good ability to dissolve lithium salts, and chemical stability with lithium metal, but oxidative decomposition hinders direct contact to CAM like nickel-rich oxides.[91,143,151–154] Further relevant examples are poly(ethylene carbonate) (PEC),[155] poly(propylene carbonate) (PPC),[155] polycaprolactone (PCL),[156] poly(trimethylene carbonate) (PTMC),[157] poly(acrylonitrile) (PAN),[86] polysiloxane (SI),[158] poly(methyl methacrylate) (PMMA),[159] polyvinylidene fluoride (PVDF),[160] and poly(bis((methoxyethoxy)ethoxy)phosphazene) (MEEP).[89,100,161] As succinonitrile (SN) exhibits a polymer-like plasticity, it is often included in this SE class or used as additive.[85,162–164] Also, single-ion–conducting polymer SE, where anions are covalently bonded to polymer backbones, are interesting, because they offer beneficial cationic transference numbers close to unity, but inherently low ionic conductivity and higher production costs are major challenges.[96] The specific properties of polymer/lithium salt complexes obviously also depend on the incorporated salt, which can be inorganic (e.g., $LiBF_4$, $LiPF_6$, $LiClO_4$, and $LiAsF_6$) or organic (e.g., $LiN(CF_3SO_2)_2$ (LiTFSI), $Li(SO_2F)_2$, CF_3SO_3Li, $LiN(SO_2C_2F_5)_2$, and $LiB(C_2O_4)_2$ (LiBOB)).[92,161] Although each salt has its own specific drawbacks, organic lithium salts are generally preferred due to high oxidation resistance, thermal stability, non-toxicity, and low sensitivity to ambient moisture.[92]

2.7. Other Solid Electrolytes

More recently, halide SE are being explored as solid catholytes, since they offer a wide ESW and reasonable ionic conductivities, but are not mostly not stable against lithium metal.[129,165–172] For example, materials with the composition Li_3MX_6 (M = metal element such as Sc, Y, In, Eu, Zr, and X = Cl, Br, I) provide net RT ionic conductivities up to 3.0 mS cm^{-1}[171] and doping with abundant metals, for example, iron $(Li_{2.25}Zr_{0.75}Fe_{0.25}Cl_6)$[173] could offer cost-effective production. Panasonics' very recent discovery of oxychlorides ($LiMOCl_4$, M = Nb, Ta) with conductivities >10 mS cm^{-1} might open up new avenues for material research, in particular because the lithium-ion density is lower than for most ceramic SE, that is, less Li is needed for SE production.[174] However, it remains to be seen whether oxyhalides with more abundant and cheaper elements than Nb and Ta can be identified. Moreover, lithium closo-borates (e.g., $70Li(CB_9H_{10})$-$30Li(CB_{11}H_{12})$)[175] and hydridoborates (e.g., $LiBH_4{\cdot}CH_3NH_2$)[176] offer superionic RT conductivities (i.e., particularly high ionic conductivities), but their poor instability toward air and moisture beside other disadvantages limit practical application in batteries.

2.8. Most Promising Solid Electrolytes and Their Development

In conventional LIB, one LE is used for the whole cell and is optimized by addition of additives addressing the specific application requirements. In ASSB, more than one electrolyte is most likely needed, and each can be tailored to the specific requirements at different cell parts. The solid catholyte needs to mitigate volume changes in the cathode, form stable interfaces toward the CAM and needs very high effective ionic conductivities for fast charging SSB cells. So far, only sulfide SE such as argyrodites and LGPS-type SE, and oxychlorides achieve very high lithium-ion conductivities, but the sulfides need protection toward oxidation, for example, by CAM coatings, and the oxyhalides contain expensive elements. To achieve very high effective ionic conductivity in the cathode composite, optimal percolation is paramount and thus, engineering of pore-free cathode composites, optimizing the particle size distribution, as well as preventing crack formation during cycling (chemomechanical contact loss) are key R&D challenges.

In SSB cells with LMA or Li reservoir free anodes, the solid separator needs to be resistive against lithium dendrite formation, chemically stable toward the lithium metal or must at least form a stable SEI, and needs sufficiently high ionic conductivities. The fabrication of pore-and crack-free separators with homogeneous

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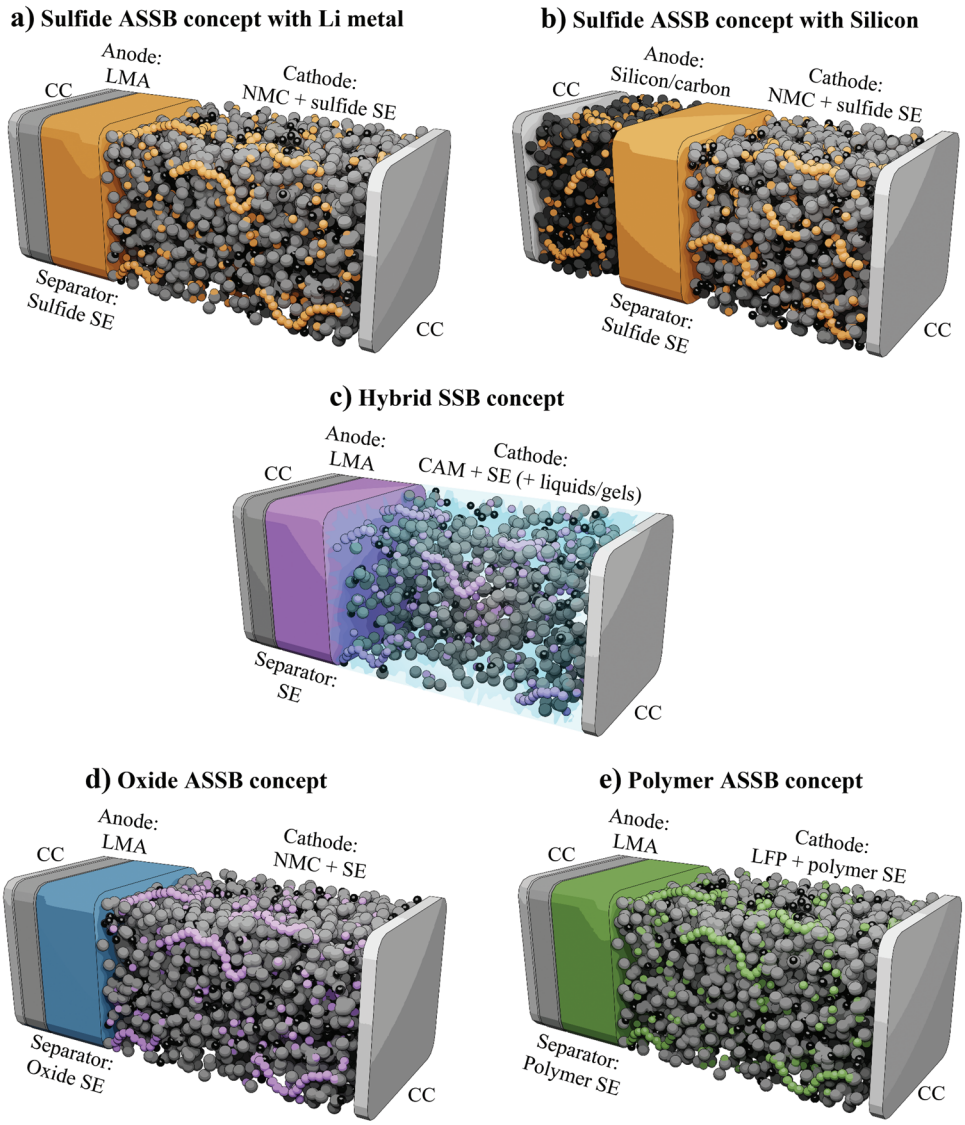


Figure 4. Graphic illustrations of promising SSB cell concepts based on a) standalone sulfide SE (orange) with LMA, b) standalone sulfide SE (orange) with silicon anode, c) hybrid setup containing unspecified SE as catholyte and separator (violet), and depicting the trend to add certain amounts of liquids or gels (turquoise catholyte), d) oxide SE separator (blue) with non-oxide catholyte (unspecified, violet), and e) standalone polymer SE (green) with LMA. (CC: current collector; SE: solid electrolyte; CAM: cathode active material; LMA: lithium metal anode; NMC: $LiNi_{1-x-y}Mn_xCo_yO_2$; LFP: $LiFePO_4$.)

surfaces is crucial. All types of SE seem feasible as separator materials, provided that they are chemically sufficiently stable versus the anode material and are dendrite-free in case of LMA. Currently, garnet-type material and sulfides, as well as polymers are applied as separator materials. Once an anode composite with silicon, graphite or LTO particles will be used, the same requirements as for the catholyte need to be obeyed for the anolyte (except of course reduction stability instead of oxidation stability). Here, garnet-type and sulfide materials dominate.

3. Promising Cell Concepts

In the following, promising cell concepts with sulfide, oxide, and polymer SE as well as hybrid cell concepts are presented.

3.1. Sulfide Solid-State Battery Concepts

Several sulfide SE exhibit a high ionic conductivity, which makes them suitable for use as both SE separator and catholyte/anolyte in composite electrodes (see section 2.3). Especially the LPS glass SE and chlorine argyrodite SE are promising candidates for sulfide SE based cell concepts. Accordingly, various systems are being developed, focusing on combinations of NMC cathodes and a silicon anode or LMA (see **Figure 4**a,b). These cell concepts promise high energy densities, but face challenges, especially with regard to the interfacial stability and lithium dendrite penetration. Prototype cells for both anode concepts have been demonstrated and industry is working on improvements on cell level and production scale-up.

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Typical composite cathodes consist of a mix of CAM, sulfide catholyte, conductive agents, and organic binders. Many sulfide SE are incompatible with NMC or NCA at high voltages[177,178] leading to detrimental irreversible reactions at the interface. Recently, strongly exothermic reactions have been demonstrated between these CAM and sulfide SE that can play a critical role for the thermal stability of sulfide-based cells.[179] CAM coatings (e.g., with $LiNbO_3$ or $LiAlO_2$) are a way to stabilize the interface,[180,181] but require an additional step in CAM manufacturing. The same sulfide SE material can be used as separating SE and as catholyte. To account for chemical compatibility toward AAM or CAM, systems with different sulfide SE catholyte and separator are also possible. Since sulfide SE separator layers of typically few to tens of µm thickness do not have "free-standing" stability, the separating SE layer can be directly coated onto the cathode. The SE layer itself then consists of dense particles or even a layer of molten/quenched SE.[182]

Sulfide SEs are thermodynamically not stable against reaction with lithium metal, leading to reduction of the SE and the formation of an SEI-type interlayer. This resulting passivating layer is electronically insulating and increases the interfacial resistance.[183] Sulfide SE containing metal ions such as $Ge4^+$ or $Sn4^+$ do not form a stable SEI and degrade much stronger.[184,185] In order to stabilize the sulfide SE against lithium metal, a lithium-ion conducting interlayer (e.g., LLZO, LiI, LiF)[186] can be applied.

Sulfide SSB are very promising for fast charging applications (see section 2.3) because of the high ionic conductivity of sulfide SE. However, in combination with an LMA high charging rates are problematic and can lead to dendrite formation[187] due to uneven current distributions. Particularly grain boundaries and pores in the SE might present channels for dendrite growth, which is why the requirements for high SE homogeneity and low defect density are very high. The reversibility of the lithium metal deposition process can be supported by external (stack) pressure on the cell or elevated operation temperatures.[21]

The high ionic conductivity of sulfides also allows the use of composite anodes. In particular, silicon-based anodes are under investigation and might allow high current rates during charging and discharging.[188] Sulfide SE anolytes might solve the problem of SEI formation and cracking that occurs with LE concepts,[189] but do not provide the good permeation and wetting by the LE and, hence, resilience to structural changes. Depending on the extent of capacity utilization, the silicon particles may experience a volume expansion of 300% or more during lithiation exerting severe mechanical stress to the electrode and the particle–particle contacts between AAM and anolyte. Respective cell concepts may therefore require a high volume fraction of 20 to 30 vol% of anolyte, and also high fractions of binder and conductive agents in the anode. The challenges associated to 3D-structural changes may be mitigated by utilizing a 2D (structured) silicon layer and hence a 2D interface to the SE.[188,190] Respective concepts have, however, lower technological maturity as compared to silicon particle composite anodes.

A number of companies are involved in the commercialization of sulfide SSB, such as SVolt, Panasonic, Samsung SDI, Solid Power, or Toyota[191–196] and have demonstrated prototype cells in pouch format. So far, these cells—in the best case—seem to be limited to less than 20 stacked electrodes. According to some communications, specific energies on cell level of around 350 Wh kg^{-1} have been reached for the LMA as well as the silicon anode concept, clearly exceeding values of state-of-the-art LIB (typically <300 Wh kg^{-1}, see cell database from ref. [197]). Fast charging was demonstrated at 2C for the silicon anode concept.[195] It is unclear whether or not automotive grade cells with a capacity of more than 50 Ah are available yet.

3.2. Oxide Solid-State Battery Concepts

Because of their (electro-)chemical stability, it is theoretically possible to combine oxide SE with most active materials, particularly at the cathode side.[198] One of the currently most promising combinations is focusing on the LMA and NMC cathodes. require the use of liquid or gel catholytes in order to achieve sufficient contact and are therefore often named as "semi-solid" or hybrid cell concepts (Figure 4c). Such concepts are in the R&D or prototyping phase of various battery manufacturers, working on their commercialization.

Due to the stability in contact with lithium metal, garnet-type SE (e.g., LLZO) are considered as promising separator materials. The production of thin and uniform layers is challenging but generally possible. In laboratory test cells, separators with layer thicknesses below 30 µm, even as thin as 10 µm, were already used.[199–201] LLZO can also be used as a protection layer, enabling the use of other oxide SE, such as LATP or LAGP,[73] or other non-oxide SE (see section 3.4). This approach can be advantageous to reach higher overall ionic conductivities.

On the cathode side, a wide range of material combinations is conceivable.[199,202,203] According to experts, oxide SE with LFP and LMA could achieve energy densities comparable to LIB with NMC. However, due to the focus on energy density, it is likely that high-nickel NMC materials will be used first. Due to the comparably low ionic conductivities of oxide SE (Figure 3), it seems not possible to operate a high-performance cell with oxide SE catholyte in the near future. Ionic conductivities >0.1 mS cm^{-1} provided by most oxide SE are sufficient for a thin SE separator layer (see quantitative limits in Figure 3), but not for the thicker layer of catholyte or anolyte. In addition, the grain boundary resistance of LLZO is high and the material is difficult to press due to its brittle properties.[204] Therefore, combinations with gel electrolytes may be used in the near future (also see section 3.4). With that, ASSB solely based on oxide SE will not be available on the market in the near future.

Some oxide SE can be combined with lithium metal without further coating.[205] This encourages the use of the LMA in combination with oxide SE because high energy densities can be achieved in this way. Different LMA concepts are possible: On the one hand, a lithium-foil electrode is conceivable, on the other hand a lithium reservoir free (see section 2.1) approach could be implemented in the future. Today, lithium-foil layer thicknesses of around 30 µm can be achieved.[206,207] Porous oxide scaffolds are also studied, however, these compromise the specific capacity of the LMA and may not be part of cell concepts for EV applications.[208–210]

According to our calculations, 320 Wh kg^{-1} and 1000 Wh L^{-1} are possible at cell level for the cell concept with gel catholyte (**Figure 5**, details on the calculation in Supporting Information).

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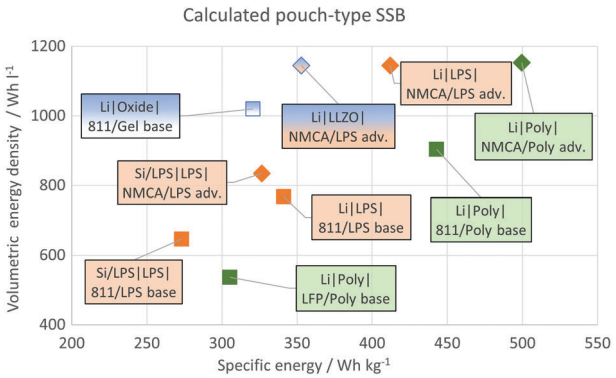


Figure 5. Calculated theoretical volumetric energy densities and specific energies of different SSB cell concepts. Details on the calculations and assumptions are described in the Supporting Information. (LLZO: $Li_7La_3Zr_2O_{12}$; LPS: xLi_2S-(100 − x)P_2S_5; Poly: Polymer SE; NMCA: $LiNi_{1-x-y-z}Mn_xCo_yAl_zO_2$;811 = NCM811: $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$; LFP: $LiFePO_4$; base: base scenario; adv.: advanced scenario, details in the Supporting Information)

Fast charging with up to 4C was achieved for oxide SSB concepts containing gel electrolytes.[211] However, this is apparently the critical current density for such concepts since lithium dendrite penetration along the grain boundaries sets in above 4C despite the mechanical stability of oxide SE.[212,213] Chemical aging has on the other hand no relevant influence on oxide SE. But mechanical impacts such as cracks can negatively influence the cell performance and, for example, lead to increased contact resistance, due to the brittleness of the oxide material. Prototype cells were reported to have a cycle life of more than 1000 cycles.[214] Oxide SSB can provide advantages in safety, as, due to the electrochemical stability of many oxide SE, the risk of thermal runaway is minimized.[137] Nevertheless, the cells contain reactive lithium metal and flammable gel electrolyte, which brings safety challenges on its own.

Industrial players like ProLogium, QuantumScape, and Qing Tao are in the commercialization phase of the technology.[57,193,215] The first production-ready cells have been announced for 2023.[215] The gel electrolyte will probably be only a transitional solution for the oxide SE. In principle, the use of oxide SE is promising because of the stability, but there are challenges in processing and especially in the associated upscaling. The possibility to further develop a hybrid concept (Figure 4d) with a sulfide SE catholyte (high ionic conductivity) together with an oxide SE separator (high stability) is being discussed and can provide advantages in terms of safety and cell performance.

3.3. Polymer Solid-State Battery Concepts

SSB based on polymer SE and LMA are already being commercially manufactured and used on a small scale (<2 GWh) in various applications (e.g., busses and stationary storage).[216] By combining the LMA and polymer SE with an LFP cathode (Figure 4e), a cost-competitive battery with a moderate energy density could be produced when scaled up.[217] The main drawback of current polymer-based battery cells is the required temperature management to maintain an operating temperature well above RT, for the sake of sustaining reasonable ionic conductivities of the SE. But even at elevated temperatures (typically 50–80 °C), relatively poor charging rates are achieved using polymer SE catholytes (e.g., C/5 to C/4 for Bolloré's Blue Solutions,[218] also cf. Figure 3), limiting the application in the EV mass market. Even though polymer SEs are commonly based on PEO, a wide range of materials and material combinations are being investigated and developed (see section 2.3)[92] and are likely to be exploited for polymer SSB in the future. While operation of the LMA appears possible, polymers are typically not stable at high potentials, which currently limits their application still to LFP and other medium-voltage CAM.

The mechanical stability and ionic conductivity of polymer SE depends strongly on the temperature and the materials used (section 2.3). Two or more types of polymers can be combined to ensure sufficiently high mechanical stability as one prerequisite for dendrite suppression, while providing a reasonable ionic conductivity, at least at elevated temperatures.[217,219,220] Furthermore, the approach of in situ polymerization of polymer SE has recently attracted attention in R&D[221,222] and industry, with which advantages in processing and interfacial contacts are claimed.

Due to the limited ESW of polymer SE, LFP cathodes are a logical choice. By mixing the polymer SE into the cathode, a good cathode–catholyte interface can be achieved.[149] The combination with higher voltage CAM, such as NMC or NCA, are so far not realized beyond the lab-scale, due to the aforementioned limitations. However, the development of new polymers such as true single-ion conductors or the use of cathode coatings can open this opportunity in the near future.[217,157,223]

The LMA is already used in polymer SSB and will most likely remain in focus for future polymer SSB concepts, as polymer SE typically show good compatibilities with lithium metal. By simultaneously using the LMA as active material and CC, the amount of copper—commonly used as CC—can be minimized. This approach reduces the inactive weight within the cell, but increases the amount of required lithium. Besides LMA, approaches with silicon anodes are investigated as well.[224]

On a cell level, theoretical specific energies of 300 Wh kg^{-1} and volumetric energy densities of 540 Wh L^{-1} could be achieved for a PEO-based lithium–metal–polymer battery containing an LFP cathode (see Supporting Information for details on the calculation). This value drops significantly on pack level, which is partly due to the heating system required for the operation of the battery. The heating system in current concepts is required to maintain the operating temperature of the battery, leading to a slow but constant drain of energy. To be competitive with state-of-the-art LIB, a low price must be achieved by this cell concept, as most other KPI are comparable. Since the production can be based on dry extrusion processes,[218,225] the need for energy-intensive drying steps is eliminated. Considering furthermore that no electrolyte filling step is necessary, and the formation process can potentially be shortened, a low production cost could be achieved. However, upscaling of these processes has yet to take place to leverage cost benefits. The low cost of LFP and PEO give further cost advantages, making the lithium price crucial for this cell concept if the lithium reservoir-free anode concept cannot be applied. The materials used in novel polymer SE vary strongly and can have a major impact on the final price.

The largest manufacturer of polymer SSB is Bolloré's Blue Solutions with two factories in France and Canada.[218]

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Polymer SEs are also incorporated in the approaches of Ganfeng Lithium Co., Hydro-Québec, Factorial Energy, Brightvolt, and Bluecurrent.[226–230]

The use of novel polymer SE, with the goal to increase the ionic conductivity at RT or allowing the use of higher voltage CAM, such as NMC or NCA, are at the center of current research activities.[149,231–234]

3.4. Hybrid Solid-State Battery Concepts

Until now, there is no ideal SE candidate, which fulfills all requirements as single electrolyte for the whole ASSB cell (compare section 2.3). As a result, hybridization concepts (Figure 4c) came into focus in R&D to effectively enhance the electrochemical performance of SSB.[61,147,235,236] In such concepts, one SE is mixed with (at least one) other SE, LE, or non-conductive additive (see below).[237,238] Different approaches are conceivable: i) cell assembly with a prefabricated composite of different electrolytes (hybrid electrolytes), ii) different SE are used as different cell components, that is, as catholyte, anolyte, and separator (hybrid ASSB), or iii) liquid additives are introduced during manufacturing of an SSB cell (hybrid solid–liquid battery cells). The taxonomy to generally describe such hybrid concepts/electrolytes is vague and inconsistent in literature with various terms being used for identical systems.[234] We advocate to distinguish at least hybrid all-solid-state and hybrid solid–liquid concepts.[93,239]

Hybrid all-solid electrolytes contain (at least) two different inorganic SE or polymer SE plus inorganic SE.[93,240] Tuning the properties of the SE by combining the advantages of different SE is currently widely pursued in research, but the actual benefit for practical application is often unclear. Thus, new issues and interfaces are introduced, and the disadvantageous properties of each individual SE can also remain. The composite can be three-dimensionally mixed, for example, polymer-in-ceramic or ceramic-in-polymer, or stacked in a layer-like concept.[241] Ceramic SE embedded in polymer SE are often referred to as active inorganic fillers.[242,243] On the other hand, hybrid solid–liquid concepts (as depicted in Figure 4c) can be regarded as intermediate concepts between LE LIB and true ASSB. This is, mixing inorganic or polymer SE (main phase) with ionic liquids (e.g., PYRxTFSI),[234,244–247] LE,[248,249] or organic liquids (e.g., ethers, carbonates, glycols).[250–252] Polymer matrices, which absorb LE as the main conducting phase, are called gel polymer electrolyte for historical reasons.[93,253,254] Due to the use of liquid or gel electrolytes, hybrid liquid–solid battery concepts can solve remaining issues, both in cell design and in production, and especially have advantages in a timely scale-up compared to ASSB concepts. In particular, the production of the cathode composite is facilitated as an intimate interface between CAM and electrolyte is straightforward with LE and gel electrolytes as they act as "contact agents" to mitigate chemomechanical issues such as detrimental contact losses, and thus avoiding unpractically high external stack pressure. For example, adding minor amounts of liquids can minimize interfacial resistance between inorganic SE and active materials.[249] This can be challenging with SE and, depending on the material, sometimes even involves problematic co-sintering steps. In addition, the increased ionic conductivity of LE as compared to many SE may offer performance advantages.[144] However, new heterointerfaces between LE or gel electrolyte and SE are formed in liquid–solid battery concepts and might cause issues in term of chemical and electrochemical stability, and in-depth analyses on the interaction of liquid additives with the solid components are necessary. For example, chemically instable interfaces between SE and conventional LE can result in the formation of resistive solid–liquid electrolyte interphases.[255] Also, side reactions between liquid additives and in particular the LMA can cause cell altering or trigger thermal runaway and thus revive the safety issues of lithium metal in LE LIB.[256] The real benefits of such hybrid liquid–solid cells as compared to state-of-the-art LIB need to be proven in terms of lifetime, fast charging capability, and safety aspects with LMA, for example, by nail penetration tests.

Many different combinations of SE/SE, SE/LE, or SE/liquid have been studied and we refer to the several reviews on this topic.[61,93,99,144,147,240,257–259] For hybrid ASSB concepts, fast ion-conducting sulfide SE are most conceivable as solid catholytes to compete with LIB for EV applications (cf. section 2.3). To suppress dendrite formation and avoid chemical side reactions at the LMA, introducing a polymer or oxide SE separator with reasonably high ion conductivity at room temperature seems currently unavoidable (hybrid ASSB cell concepts). For example, an LPS catholyte could be used in combination with the LMA if protected by an LLZO separator in a bilayer hybrid SSB cell since the interfacial resistance between these SE turned out to be very low.[260] Such a cell (LMA|LLZO|LPS|NMCA) could deliver 355 Wh kg^{-1} and 1150 Wh L^{-1} at cell level according to our calculations (Figure 5). Many R&D activities also pursue combinations of polymer SE (mainly PEO-based) with oxide or sulfide SE such as LLZO, LGPS, LAGP, LPS, or argyrodites.[61,261,262]

If flammable liquid components are introduced to the SSB cell, safe operation with LMA has to be proven. Apart from that, an inherent advantage of hybrid SSB concepts is that, with the right choice of electrolytes, any active material at the electrodes is conceivable. As a prerequisite, the (electro-)chemical stability between the different SE or liquids must be assured.

Several of the companies mentioned in sections 3.1 to 3.3 also focus on hybrid SSB cell concepts with the aim of timely commercialization, for example, Ganfeng Lithium Co.,[263,264] QingTao,[264] WeLion/NIO,[264,265] ProLogium,[264] Brightvolt,[266] QuantumScape,[57] Bluecurrent,[230] or Factorial Energy.[267] In fact, some SSB mass production announcements are based on hybrid solid–liquid concepts, but the benefits of such batteries is an unsettled matter concerning a safe and reliable long-term operation with LMA in comparison to ASSB and in terms of energy and power density as well as costs in comparison with state-of-the-art LIB.

3.5. Conclusions on Cell Concepts

Diverse cell concepts have been proposed and their number is high. An interpretation of industry activities in particular helps to suggest which concepts, among others, are the most promising. As shown above, major industry activity takes currently place in the field of hybrid concepts. As an intermediate development step toward ASSB, it is likely that this technology will reach larger production capacities first.

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As for ASSB, advantages exist for each cell concept and the use case and application requirements will decide, as well as technical and production aspects, for the success of one or the other concept and their temporal implementation (see also section 5.4 and Figure 7). As of today, no explicit favorite of the cell concepts can be named.

4. Solid-State Battery Production Aspects

SSB differ in materials as well as construction from state-of-the-art LE LIB. While some steps during cell production are likely to be transferable on SSB, others differ significantly and require new processing technology. An overview about current and future processes is given in **Table 1**.

4.1. Electrode Composite Processing (Cathode, Anode)

In the most promising SSB cell concepts (section 3), cathode and anode (only in case of silicon- and graphite-based anodes) consist of composites of active material (CAM and AAM), SE, conductive additives, and binders. In line with the state of the art in LIB production, wet processing and potentially dry processing techniques can be used for electrode production. Wet chemical processes based on *N*-methyl pyrrolidone or water combined with fluorinated binders or styrene–butadiene rubber (SBR) enable a high throughput. These solvents are compatible with most oxide SE and polymer SE. However, some sulfide SEs are not stable toward polar solvents and replacement by non-polar solvents, for example, xylene seems unavoidable for these SE.[206,268,269] This would, however, require the development of new binder systems, since commonly used PVDF is practically insoluble in non-polar solvents. SBR and silicon rubber (SR), on the other hand, show compatibility. For high silicon content or silicon standalone anodes, there may be a need for specialized binders which can withstand the high mechanical stress caused by silicon particle volume change during de-/lithiation.[270–272] The instability of sulfide SE with water also leads to additional requirements for an extremely dry or even inert processing atmosphere in all steps where the SE is handled in powder form.

The slurry coating and drying process might be comparable to LIB electrode production. It could, however, have to be repeated multiple times to create layered structures of cathode or anode composite, interlayers, and separating SE layer. Similar to LIB production, the coating process is followed by electrode densification to enhance particle–particle contacts.

If a malleable sulfide SE is used, a calendering step similar to state-of-the-art LIB processing might be sufficient. SE layers based on oxide materials might require a sintering step to ensure low porosity and low grain boundary resistance in the electrode. The sintering step will probably increase the energy footprint and decrease the throughput of electrode manufacturing. In combination with typical CAM, sintering temperatures are limited to about 700 °C, below the decomposition temperature of the CAM.[273,274] Alternative approaches such as melt infiltration of the SE into a porous cathode structure are investigated, since they might omit an additional sintering step.[275]

Wet processing routes show the highest market maturity for sulfide and oxide SE, although particularly the electrode drying is energy intensive and costly.[276] Therefore, the long-term trend points toward dry processing, or wet-processing

Table 1. Comparison of different cell concepts and manufacturing processes from electrode manufacturing to cell assembly.

Production	LIB	Sulfide SSB	Oxide SSB	Polymer SSB	Long-term goal
Anode	Wet processing Slurry mixing and coating, drying, calendering	Extrusion process (Li foil) Extrusion, calendering, lamination Wet processing (Si-based anode) Slurry mixing and coating, drying, calendering	Extrusion process (Li foil) Extrusion, calendering, lamination	Extrusion process (Li foil) Extrusion, calendering, lamination	Melt or vapor-based process (Li foil) Simplified dry or wet processes for host-structure deposition for Li reservoir-free anodes
Cathode composite	Wet processing Slurry mixing and coating, drying, calendering	Wet processing Slurry mixing and coating, drying, calendering	Wet processing Slurry mixing and coating, drying, low-temperature sintering	Extrusion process Extrusion, calendering	Dry processes or green solvent-based processes
Separator	Roll-to-roll Dry/wet extrusion process (PP/PE) and roll-to-roll feeding in cell assembly	Wet processing Slurry mixing and coating, drying, calendering	Wet processing Slurry mixing and coating, high-temperature sintering, lamination, low-temperature sintering	Extrusion process Extrusion, calendering	Dry processes or green solvent-based processes
Cell Assembly	Cell-assembly Stacking/winding, packaging, electrolyte filling, formation, degassing, aging	Cell-assembly Electrode stacking and stack pressing, packaging, (shorter) formation and aging	Cell-assembly Electrode stacking and stack pressing, packaging, (shorter) formation and aging	Cell-assembly Electrode stacking and stack pressing, packaging, (shorter) formation and aging	Omit formation and aging; SSB round cells; SSB production with low CO_2 footprint

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techniques with only little amount of (ideally green) solvent. Polymer SE are already being processed by solvent-free extrusion on an industrial scale.[218,225] Other processes like dry printing or electrostatic spraying are investigated for other SE-types.[187,277]

4.2. Lithium Metal Anode Processing

There are different processes to produce lithium metal films, for example, extrusion, melt-processing, and vapor-based processing. The dry-extrusion of thin lithium films followed by repeated calendering is currently the most established technology.[206,273,278] Thin layers with a thickness of <30 µm and high homogeneity are considered challenging.[206,207] In particular the adhesive properties of lithium metal render it hard to process and require specialized calendering technology.[278] Already at a thickness of 50 µm, the electrode cost significantly exceeds the lithium metal cost.[273,279]

The demand for thinner foils has led to the development of other techniques, such as melt processing, where lithium is deposited onto a substrate in liquefied form.[206,280] The method is especially applicable for porous substrates, for example, separators or structured CC. However, it is not yet industrially available and requires conditions like vacuum or inert gas atmosphere which are not common in electrode production. Extremely thin lithium layers of a few µm can also be produced by vapor-based processing, such as sputtering.[206] But again, the feasibility for mass production of high-capacity battery cells has not been demonstrated yet.

To avoid costly production and assembly of initial LMA layers and potentially reduce the amount of lithium needed, lithium reservoir free (see section 2.1) cell concepts are in the focus of several research activities.[21,207,281] Current approaches mostly use tailored CC or host structures,[21] however, which causes an additional step in CC or cell production.

4.3. Solid Electrolyte Separator Processing

Similar to the processing of composite electrodes, wet and dry coating techniques can be used to coat the separating sulfide or oxide SE layer. Typically, this can be done by application of a second coating onto the cathode or anode and results in a layer thickness of 5 to 10 µm separating SE.[269] Alternatively, the separating SE can be manufactured as a free-standing SE.[187,269] Respective SE layers need to be highly mechanically stable, which often requires a certain minimum thickness for inorganic SE (>30 µm) or the use of supporting organic binders, which enhance the interconnectivity of inorganic SE particles, but may lower the ionic conductivity and lithium dendrite resistance.[268,269]

With both approaches, the layered structure of electrodes and separating SE needs to be densified either by calendering, pressing, or by sintering, to ensure good contact, low porosity, and high ionic conductivity. The potential of mechanical densification is limited by the ductility of SE materials.[206] Particularly for polymer but also for some sulfide SE, a sufficient density can be reached by calendering. Most oxide SE cannot be compacted sufficiently by purely mechanical methods and require sintering. In the case of separate production processes for a free-standing SE layer and the cathode, two sintering steps and hence different sintering temperatures can be applied, which would allow to achieve a high density in the separating layer while avoiding CAM decomposition due to high temperature treatment at the same time. Other approaches utilize sintering agents, such as Al_2O_3 and Li_3BO_3[282,283] or ultrafast high-temperature sintering, for example, by resistive heating techniques.[284]

4.4. Cell Assembly

Slitting of the electrode sheets and stacking may follow similar steps as for state-of-the-art LIB. However, the thickness of combined electrode and separator layers as well as lithium metal as a new material in the process may be challenging for the cutting process. For the stack assembly, a heating and pressing step might be beneficial for many SSB concepts to provide good interface contacts between the individual layers in the stack.[206,273] Unlike for LE LIB, the electrolyte filling and degassing steps can be omitted for SSB concepts without any LE. It is unclear, how typical formation and ageing procedures for SSB will look like, however significant time saving can be expected compared to LIB, in the long term.[278,285] The formation might potentially be even omitted completely for SSB with concepts using an initial LMA.[206,278,285]

5. Roadmap

5.1. Key Performance Indicators of Solid-State Batteries

The research and development of SSB is motivated by the potential improvements of key properties (performance indicators) with respect to state-of-the-art LIB. However, which improvements are actually feasible depends heavily on the components used.

SE are often mentioned when improvement of battery safety is discussed. SE have in general a substantially lower flammability than the LEs used in conventional LIB.[286,287] Furthermore, due to the absence of liquids, no leakage can occur, reducing the risk of contamination.

The adoption of an LMA comes with many safety risks, which can be only partly mitigated by the SE: for most LMA concepts, the formation of dendrites at high current densities has been reported.[288] If this problem cannot be solved by the design of the anode, higher densification of the SE could be a possible solution. The mechanical stability of an SE is one vital parameter in the complex interplay of factors affecting the resistance to dendrite formation (see section 2.1) and crucial for the overall safety of the SSB.[289]

Polymer SEs tend to have lower mechanical stability than ceramic SE and in addition, often exhibit too low ionic conductivity at RT for fast-charging applications. Sulfide SE can form the toxic and inflammable gas H_2S, when in contact with water, posing an additional safety challenge.[290] Corrosive SO_2 may form at high

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charging potentials, once the sulfide catholyte is not sufficiently protected against reaction with the oxide CAM.[291] The low reactivity and high mechanical stability of oxide SE make them favorable in terms of safety properties. Recent thermodynamic models indicate that short-circuited SSB can reach temperatures significantly higher than conventional LE LIB, indicating a potential safety issue.[256]

The achievable energy density and specific energy on cell level are closely linked to the composition of the electrodes. The adoption of LMA and nickel-rich NMC or other higher voltage cathodes is therefore a common approach for SSB. Most polymer SE cannot be paired straight-forward with CAM with potentials larger than those of LFP. Coatings of higher voltage CAM are also required to prevent sulfide SE from decomposing at the cathode interface. The layer thicknesses have a direct influence on the energy density of the cell, favoring thin electrolyte layers. To give an estimation on the volumetric energy densities and specific energies that could be reached, theoretical values were calculated under certain assumptions (details described in Supporting Information), resulting in the values shown in Figure 5.

To achieve short charging times (high C-rates) and high power density, high lithium-ion conducting SE and high quality of the cell component's interfaces are necessary. The manufacturing of low-impedance interfaces is especially challenging for the less flexible inorganic SE.[292]

The absence of liquids could improve the calendric lifetime of SSB over LE LIB, by reducing the degradation effects during battery rest. The effects limiting the cycling lifetime of conventional LIB, such as lithium plating and cathode decomposition,[293] can play a similar role in the lifetime of SSB. The decomposition of the electrolytes during cycling is especially challenging for sulfide SE and depends for polymer SE strongly on the cathode used.[294]

The price of the battery will be most crucial when it comes to the competition with conventional batteries on a large scale. Since most SE are not yet produced in large quantities, the final price of these components is hard to estimate and depends strongly on processing and material aspects. The amount of lithium used in the SE can be a main driver for the cost, possibly giving a price advantage for polymer SE.[4] The sintering needed in the manufacturing of oxide SSB is an energy intensive process, driving up the production costs. Even though dry rooms are needed for all SE classes, some sulfide SE candidates are very sensitive to water and need argon atmosphere for longer handling and storage time.[112,295] Their processing in dry rooms needs to be limited to the shortest time possible and should occur at low temperatures to suppress reactions in contact with water. Processing steps could also be transferred to microenvironments with argon atmosphere, which enhances the overall costs and hampers large scale production.

Depending on the choice of technology, SSB can compete with or even outperform conventional LIB in each important key property;[34] however, it seems unlikely that a "one-fits-all"-solution that is competitive regarding all discussed KPI will be developed in the near future, as the approaches to optimize one parameter are usually accompanied by trade-offs in another.

5.2. System-Level Aspects and Applications of Solid-State Batteries

5.2.1. System-Level Aspects

For most applications, the battery cells need to be assembled to battery packs. These packs contain the battery management system, temperature regulation and safety features suitable for the respective application. Some of the requirements on pack level are likely to change when replacing conventional LE by SE for SSB cells. As SSB will typically contain silicon-based anodes or the LMA, high volume changes during de-/charging will occur in the battery cells.[296,297] These volume changes need to be compensated by external pressure, as good solid–solid interfaces in the cell are crucial for the battery to function properly.[298] To meet the same requirements as state-of-the-art LIB, stack pressures for SSB should stay well below a few 0.1 MPa.[4] In most studies especially on laboratory cells, however, much higher values are often used.[4,298,299] While a minimum pressure is required to ensure good contact between the electrode active materials, the SE, and the CC, a too high pressure can cause cracking and thus also lead to malfunctioning of the cell.[299] Hence, a relatively low stack pressure (likely between 0.1 and 1 MPa) is realistic to be applied and is also considered as technically possible from the perspective of the battery industry and OEM.

Considering the temperature management system, less stringent requirements are probably needed for most SSB, as they can often sustain higher operating temperatures.[300] On the other hand, polymer SSB require an operating temperature of 50–80 °C and thus a heating system. Finally, safety aspects around the LMA and sulfide SE must be considered.

5.2.2. Applications

Electromobility and especially automotive applications are the main drivers for SSB development and the envisaged main application area. Polymer SSB is the only one of the three classes discussed herein that is already on the market on a larger scale, currently primarily used in electric busses.[56] The main drawbacks of this technology are the need for heating and the rather low charging rates (typically clearly below 1C), limiting the use cases to applications in which the battery is more or less continuously, or at least very regularly, in use and fast-charging is not required (e.g., industrial applications, such as automated guided vehicles). Sulfide SSB might enter the market in the consumer segment, before being implemented in the larger scale in electric cars between 2025 and 2030, as in the automotive sector long periods of testing, qualification, and product design are required, whereas the consumer sector is much faster in this regard. In the long term, various application scenarios for sulfide SSB are conceivable, such as commercial vehicles (e.g., trucks) and passenger aviation. Similarly, the main application sector for oxide SSB will be electric vehicles and the market implementation is expected in the same time frame. Due to their potential robustness, industrial and heavy-duty applications might also be conceivable for oxide SSB. Stationary applications might be possible in the long term for all types of SSB, however, only if costs per charging cycle that are competitive with LE LIB or emerging alternative

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battery technologies (e.g., Na-ion batteries) can be achieved, as the stationary sector is most price-sensitive.

5.3. Market Developments of Lithium-Ion Batteries and Solid-State Batteries

The growing global battery demand is currently being driven primarily by the expected market for EVs. Other markets such as consumer electronics and stationary storage are enhancing this fast growth in demand. The total battery market growth has been over 30% year-over-year, in recent years.[301] In 2022, the demand for LIB was ≈780 GWh. In 2023, the demand might increase to 1 TWh. By 2030, the global demand for LIB could reach about 3.5 TWh per year (market studies specify a range between 2–4 TWh).[302–308] Even after 2030, the market will continue to grow. By then, new applications, for example, passenger aviation and others, could reach a relevant market share, which will further increase the overall demand. In the long term, global battery demand of more than 7.5 TWh per year is considered realistic.[303]

It is very difficult to predict the future market share for SSB. Their demand for mass applications primarily depends on the future price developments, especially for the lithium price, which cannot be predicted with certainty. Current trends toward LFP cell chemistries in the automotive sector confirm the strong focus on price. Respective batteries can be seen as the benchmark for any technology aiming to target mass markets.[309] It is quite possible that oxide and sulfide SSB cells will have initially higher prices compared to LIB when they enter the market, due to an initially lower scale production volume and less experience in SSB production compared to LIB and a supply industry that has yet to develop. Two scenarios are conceivable for the market entry of SSB: in the first scenario, a straight giga-scale-up of SSB and thus factories for the use of SSB in automotive mass markets could generate competitive costs. Another possibility would be an initially smaller scale production with a higher price, but high technological value for niche applications, for example, due to good safety properties or high application requirements on energy density.

Since 1996, over 1.5 billion $ were invested in the industrial development of SSB.[301] Still, experts estimate the SSB production capacity in 2022 at <2 GWh. Relevant increasing cell capacities will only be added once cell concepts have reached commercial maturity and clear application areas and use cases were identified. This case could occur by the end of the decade. On the basis of forecasted technological developments of the individual SE materials, experts came to an estimate of the production capacity to be built up in the foreseeable future (**Figure 6**, details in Supporting Information). Polymer SSB production capacity may reach 2–15 GWh in 2025 and 10–50 GWh by 2035. Sulfide SSB may reach 0–5 GWh production capacity in 2025 and 20–50 GWh by 2035. Oxides could reach 0–1 GWh in 2025 and 10–20 GWh by 2035.[1] It can be assumed that the expert assessments are rather conservative. There is already a production announcement from one company alone, which has announced 100 GWh of hybrid and ASSB cells.[265]

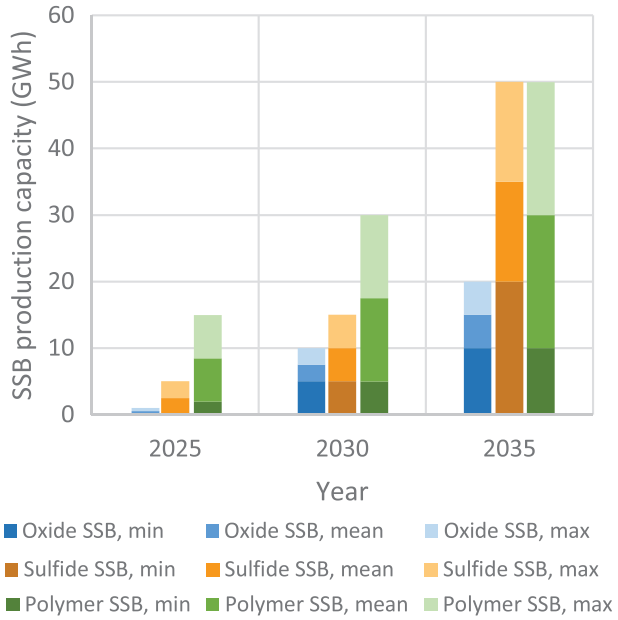


Figure 6. Estimations on SSB production capacity, as forecast by battery experts from industry and academia at a workshop in 2021 (for details see the Supporting Information).

5.4. Roadmap

Figure 7 summarizes a scenario at which time certain SSB cell concepts could enter the pilot production phase. While polymer SSB (with LMA and LFP, operating at elevated temperature) are on the market already (see section 3.3), sulfide SSB (with silicon anode) and oxide SSB (with gel catholyte) are expected to enter the larger scale pilot production shortly after 2025. Polymer SSBs with NMC are expected between 2025 and 2030, slightly before sulfide SSB with LMA. The values for specific energy and volumetric energy densities of future SSB cell concepts in Figure 7 are according to our calculations (see also Figure 5) and the temporal positioning is according to an assessment of battery experts at a workshop in 2021 (details see Supporting Information).

When comparing the anticipated energy densities and specific energies of these SSB concepts with the anticipated development of these KPI of LE LIB, it becomes clear that state-of-the-art LIB are a moving target that will improve continuously in the next years and that SSB are not likely to represent a quantum leap in performance improvements, but rather will be the evolutionary next step forward toward improved battery performance.

6. Conclusions

SSB can be considered as a further evolutionary development of LIB. Mainly driven by the demand for higher energy and power density (specific energy and power) and improved safety—nevertheless at low costs—the "solidification" of cells is explored as one of the few options for continued optimization of the LIB cell concept. Thus, any solid-state concept should not be seen as a revolutionary step, but rather as logical attempt for further improvement of an otherwise already successful technology.

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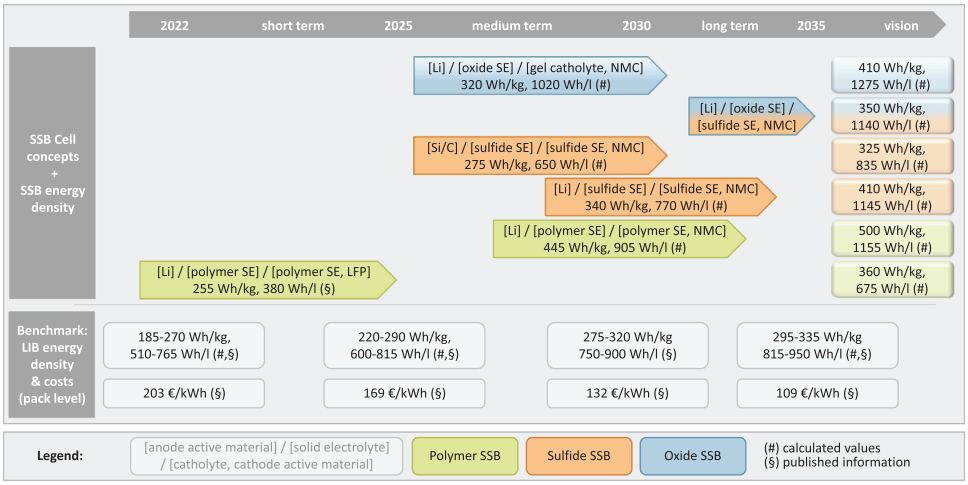


Figure 7. SSB roadmap with cell concepts, their anticipated start of large-scale pilot production and corresponding volumetric energy densities and specific energies, compared with the benchmark KPI of LE LIB (# values we calculated under certain assumptions, details are described in the Supporting Information; § polymer SSB energy density and specific energy according to ref. [225], LIB energy densities and specific energies according to and interpolated from values from ref. [310], and LIB costs according to ref. [311]). (SE: solid electrolyte; NMC: $LiNi_{1-x-y}Mn_xCo_yO_2$; LFP: $LiFePO_4$.)

This becomes particularly obvious when considering the active materials that define the maximum energy that can be stored per mass or volume of cell—which of course also depends on the sum of all inactive materials and cell housing. The same cathode materials will most probably be used in SSB and LIB, which means that the catholyte of a high-performance SSB has to operate stable and reversibly with typical layered oxide CAM or LFP. SSB can only compete with LIB in terms of specific energy (as well as energy density) once the lithium metal or high-capacity silicon anode can be utilized reliably—which is still an open question. Recent work on LIB with LE and an LMA demonstrated optimized single cells with about 700 Wh kg^{-1} specific energy, however, only for a very small number of cycles.[312] This highlights the perspective of high-capacity anodes, but we assume that only SE will ultimately allow their safe operation. In the medium term, this might be achieved by solidifying parts of the electrolyte or by solid protective layers, which brings hybrid solid–liquid battery concepts into R&D focus. In general, there are still considerable uncertainties about the best suited materials combinations and processing technologies, which reflects the yet not achieved maturity of the technology. Various industrial players seem to have found their individual concept and future will show, which of these will deliver the best performance. By and large, major R&D activities are still necessary, especially toward higher TRL, pilot production, and uptake by industry. Especially, KPI such as costs and sustainability are yet not well specified, and it is unclear if SSB will be able to offer improvements over LIB in these respects in the long term. At least in the years of market entry SSB are anticipated to be more expensive than LIB, which for their part have already been thoroughly optimized for decades. This is mainly due to the initial high development costs for new SSB process equipment and production lines, as well as tailored battery materials such as the electrolytes and coated active materials. In other KPI, in particular energy density, specific energy, fast charging ability, safety, and perhaps even stability (cycle and calendric life), SSB promise improvements. However, general target values cannot be defined for the whole group of technologies and will depend on the specific application scenario. Nonetheless, SSB still need to prove their competitiveness or even superiority over LIB on the market in the next decade, in order to achieve fast market penetration. A good mix of the most important KPI above will be necessary for SSB to succeed. As LIB are still further improving in these KPI as well, the competition between SSB and LE LIB will get more intense continuously. The industry is now moving fast, with announcements for a production capacity of almost 300 GWh (Table S3, Supporting Information). Although the timeframe is often specified, the technology is not always clear (ASSB, semi-solid–state battery, and condensed battery) and likely not all announcements will become reality. Furthermore, not all companies will announce years upfront their planned production capacity of SSBs, so the 300 GWh can be only considered a starting point and an indication that SSBs are quickly moving toward production.

In conclusion, there are strong R&D activities worldwide, with most established and large-scale industrial players in Asia, and with numerous startups in North America. Research activities are also widespread in Europe, however, here we see much fewer industrial activities.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

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