



# Insights Into Scalable Technologies and Process Chains for Sulfide-Based Solid-State Battery Production

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The successful utilization of innovative sulfide-based solid-state batteries in energy storage hinges on developing scalable technologies and machinery for upscaling their production. While multiple Gigafactories for lithium-ion batteries are already operational worldwide, the upscaling of solid-state batteries exhibiting their full potential remains to be seen in the near future. In this study, the conventional production of lithium-ion batteries is reconsidered, and the feasibility of seamlessly integrating sulfide-based solid-state batteries into the existing process chains is discussed. Scalable technologies and key

challenges along the process chain of sulfide-based solid-state batteries are accordingly addressed. Experimental investigations yield crucial insights into enabling large-scale production of sulfide-based battery components while highlighting remaining challenges from a production perspective. An overview of the roll-to-roll machinery housed in microenvironments under an inert atmosphere in the "Sulfidic Cell Production Advancement Center" at the Institute for Machine Tools and Industrial Management at the Technical University of Munich is given.

## Introduction

Research on sulfide-based solid-state batteries (SSBs) has made significant progress over the last five years aiming for energy densities similar to or higher than lithium-ion batteries (LIBs).<sup>[1]</sup> Advantages, however, remain to be proven since current SSB technologies still need to show the expected breakthrough to offer safe, cost-effective, high energy and power density cells for the broad market.<sup>[1,2]</sup> The industrialization of new battery technologies takes time, as it also took decades of work to implement the LIB in the automotive industry.<sup>[3]</sup> As the battery market undergoes a massive transition towards more cost-beneficial materials such as LiFePO<sub>4</sub> (LFP)<sup>[4]</sup> or sodium-ion batteries,<sup>[5]</sup> the near-future competitiveness of SSBs is uncertain. Moreover, numerous Gigafactories are set up worldwide and will offer production capacities in the terawatt-hour range of battery cells for mainly automotive customers. Whether additional SSB Gigafactories will be built is arguable.

More time is needed to develop SSBs – this becomes apparent from literature reviews,<sup>[6]</sup> which mainly focus on

current and future challenges instead of presenting reliable cell data from long-term cycling cells.<sup>[1,7]</sup> Since the lengthy development times do not match the rapid pace at which Gigafactories are built up, the need for an SSB technology that can "drop in" to LIB production lines becomes increasingly important.

This study discusses the applicability of LIB production processes and technologies along a scalable process chain for sulfide-based SSBs. Process chains are highlighted considering changes, which will be necessary to implement sulfide-based materials on LIB production machines successfully. Every process step is detailed and underlined with examples from experimental results and key enablers for industrial production. The strong linkage between cell components and process parameters will be in focus from a manufacturing point of view. Finally, the newly set up "Sulfidic Cell Production Advancement Center" (SPACe<sup>-</sup>) at the Institute for Machine Tools and Industrial Management (*iwb*), where the goal is to produce sulfidic SSBs in a roll-to-roll process on pilot-scale machines, is presented.

## Sulfide-Based Solid-State Battery Production

The production of sulfide-based SSBs requires an in-depth understanding of the mechanical structure and its influence on electrochemical components.<sup>[1,8]</sup> Similar to LIBs, the component and cell design of sulfide-based SSBs is a crucial enabler for achieving high-performance cells during the early stages of the process chain, where interfaces between components can be selectively modified.<sup>[9]</sup> Conventional LIBs follow a well-understood process chain where each technology fulfills one unique process step, e.g., a coating machine applies material on a substrate to form a coated layer, or a calender is used to compact a coating.<sup>[10,11]</sup> Research has shown that this clear categorization does not necessarily apply to SSB production.<sup>[12a,b,2,12c,d]</sup>

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In sulfide-based SSB production, component production can be combined with a partial cell assembly in contrast to the typical LIB production (cf. Figure 1). The terms single- and multi-component fabrication are introduced in Figure 1 to allow the definition of a new process chain with different sequences and using other processes in contrast to LIB production. The mixing step is the process of forming the dry or wet mixture of battery components at the material level. The coating, compacting, laminating, and cutting process steps can fulfill a production task at either the single- or multi-component level with multiple possible sequences. For example, the coating step can be used either for the single electrode and separator processing or to partially assemble cells by applying a second coating on top of an existing one. A cutting step can occur before compacting if a discontinuous technology (e.g., uniaxial, isostatic pressing) is applied instead of a roll-to-roll process. As a result, the intermediate products after each process step can differ. The input for a single-layer cell stacking can either be one single sheet or a combination of up to five sheets, which may include current collectors, electrodes, and a separator. The process chain ends with stacking, contacting, packaging, and finishes on the cell level.

In the following, the considered components for the production are set to a composite cathode, a sulfide-based separator and a silicon anode. While the use of other cathode active materials does not have a major influence on the process chain, the choice of anode material will determine feasible technologies. In literature, two further promising anode concepts include the use of pure lithium metal on copper foil<sup>[13]</sup> and the anode-free concept, where lithium is solely plated from the cathode to the anode side onto bare copper foil during charging.<sup>[14]</sup> These two concepts bring major variance into the production of sulfide-based SSBs, since the cell design change comes alongside challenging technologies, which are not yet mature in terms of scalability. Therefore, the following suggestions are concentrating on the silicon anode.

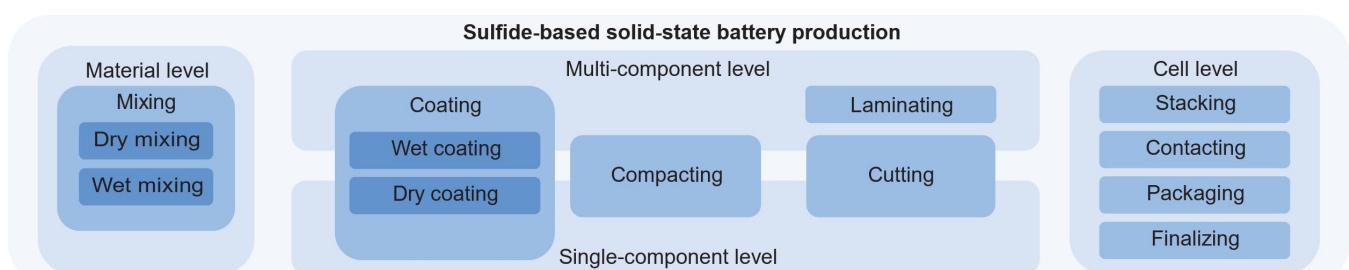
### Industrial Scale Production Chain

In Figure 2, two suitable process chains for sulfide-based SSBs are depicted in detail. In Figure 2a) a continuous process chain consisting of conventional LIB technologies is shown for the

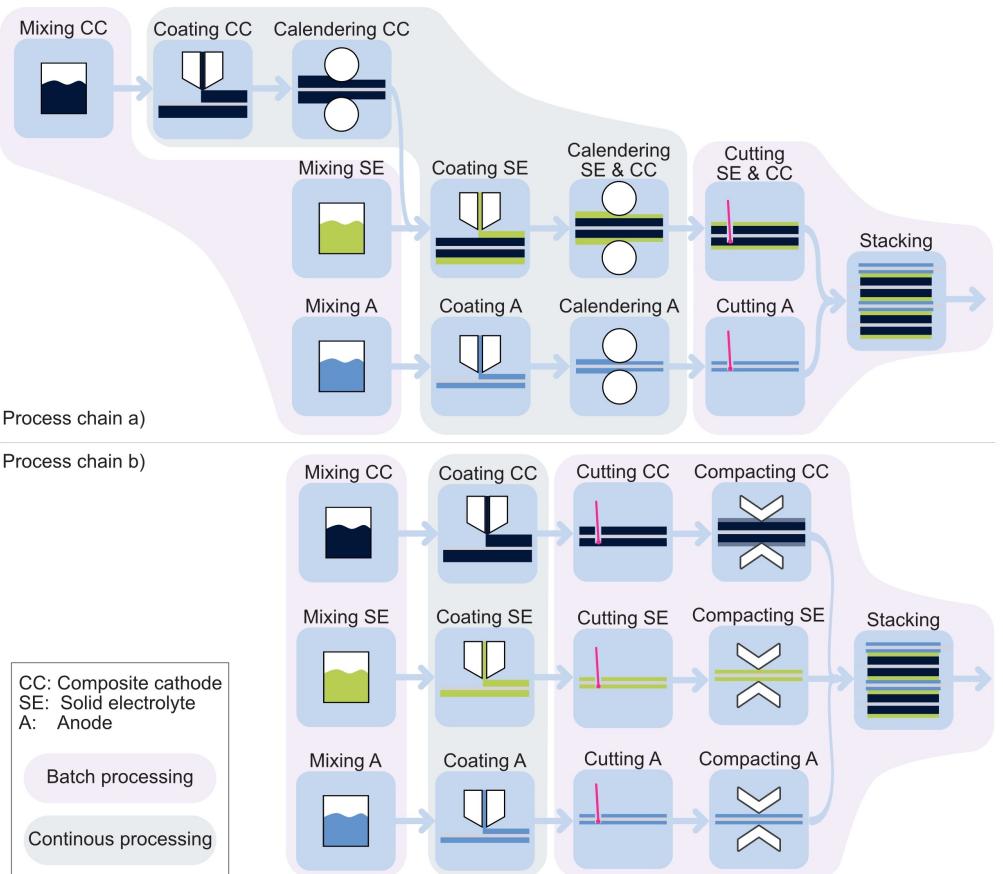
components of sulfide-based SSBs with a silicon anode. The proposed process chain begins with the production of a double-sided composite cathode analog to LIB production. A wet coating step is used to apply a slurry onto a current collector. Afterwards, the dried coating is calendered to achieve a dense electrode using roll-to-roll processes, therefore ensuring scalability and compatibility with LIB equipment. Similarly to the composite cathode, a SE slurry is prepared but coated directly on the calendered composite cathode to ensure a larger interfacial contact area between the cathode and separator compared to a combination of both single-components further down the production chain. The cathode-separator multi-component is then calendered to compact the separator layers. The multi-component is produced symmetrically, meaning both cathode and separator are coated double-sided, which is required for the subsequent calendering step. The calendered multi-component is then cut using, e.g., a laser beam process. The silicon anode proposed in the shown production chain is manufactured using a typical water-based route<sup>[15]</sup> and cut into sheets. The silicon anode production is similar to the state-of-the-art silicon blend graphite anode production used for LIBs. After the (multi-)component production, the anodes and the separator-cathode multi-component are combined in a single-sheet-stacking process. The cell finalization steps are analogous to LIB production, including tab welding, packaging, and cell finalization.

The bottleneck of the process chain shown in Figure 2a) is the cell design. Coating the separator on top of the cathode with a subsequent cutting process entails the separator being as big as the cathode with no overlap on at least two sides. In addition, the cut edges must be sharp and clean to prevent a downstream short circuit. A solution for this challenge could include the intermittent coating of cathodes, coated afterward with a separator. This would lead to the possibility of having a larger separator than composite cathode due to the gaps in between. Moreover, the coating of the SE separator on top of the anode is a solution. Again, SE solvent compatibilities with the anode need to be investigated.

In Figure 2b), a single-component process chain that involves new technologies applicable to sulfide-based materials is pictured. In contrast to the multi-component production shown in a), the electrodes and the separator are produced in parallel, independent process chains. Here, cutting has to occur



**Figure 1.** Overview of an innovative approach for redefining the production process of sulfide-based solid-state batteries (SSB). The terms single-component level and multi-component level bridge the conventional separation of electrode production and cell assembly of LIBs, as there is no clear distinction between these process steps for SSBs.



**Figure 2.** a) Example of a multi-component production for sulfide-based SSBs using common technologies to produce the composite cathode (CC) and the solid electrolyte (SE) separator as a multi-component by coating. Continuous roll-to-roll technologies are mainly applied. An anode (A) is separately fabricated using LIB technologies and stacked at the cell level. b) Example of a single component production for CC, SE, and A implicating innovative compaction technologies.

before densification because of the discontinuous nature of the applicable pressing technologies (uniaxial or isostatic). After cutting, the densification of a single-component is executed. Depending on the cell design and technology, simultaneous densification of the cathode and separator is possible (cf. Figure 2b).

The bottleneck of this chain is the high handling effort. Producing and handling single sheets in final dimensions requires precise automation and is susceptible to errors. The larger the single-sheets are, the more fragile they get, especially the SE separator. In addition, lead times will become significantly longer compared to roll-to-roll processes. A solution could be the lamination of the composite cathode and separators after single component coating.

These two process chains are exemplarily shown and are not the only options, but based on experimental results, they seem promising and feasible concerning “drop in” to LIB production lines to a certain extent. Specific details about laboratory results using the above-mentioned process steps are discussed in the following.

## Insights Into Scalable Technologies

In the following, the processes along the production chain are discussed in detail, showing experimental results and key findings underlined with expert knowledge based on five years of experience in manufacturing LIBs and SSBs.

### Mixing

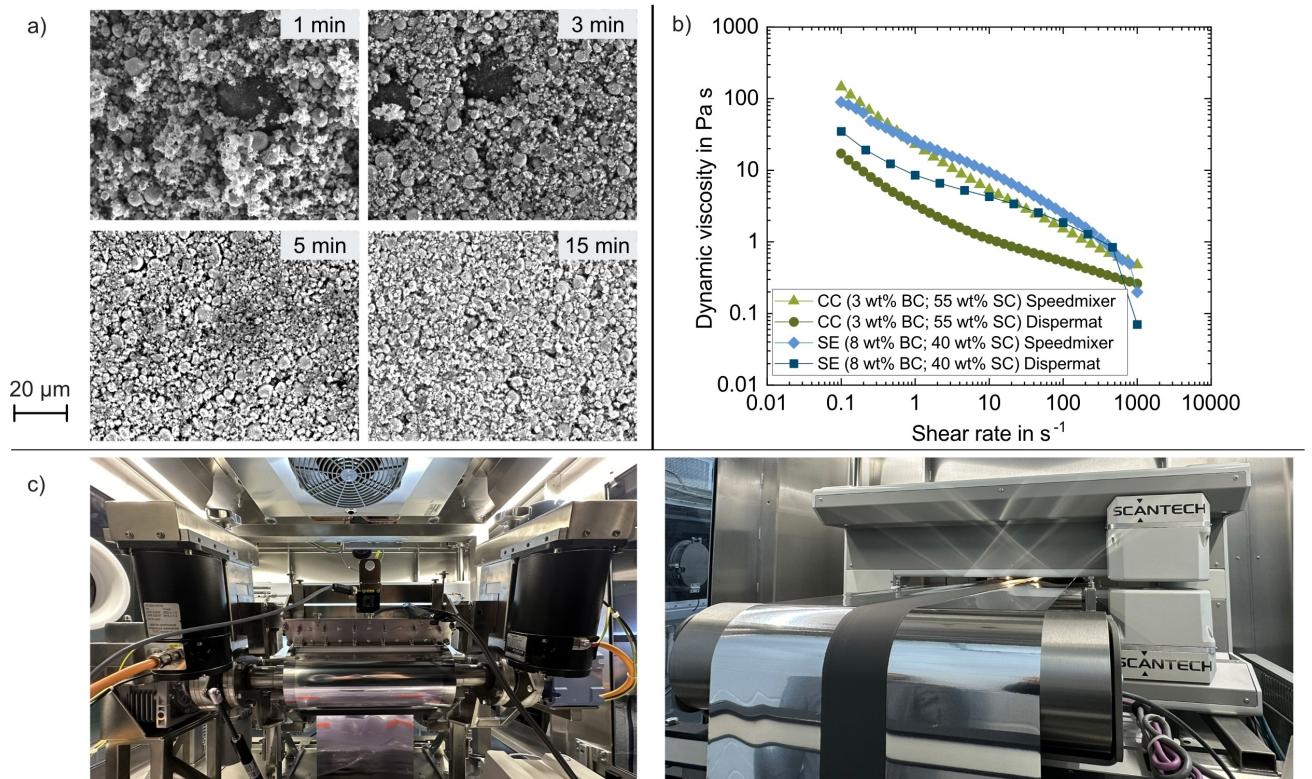
The first value-adding step for SSBs is comparable to LIB production.<sup>[11]</sup> In the mixing step for composite cathodes, the distribution of SE is crucial for achieving good and homogeneous ionic percolation pathways throughout the cell.<sup>[16]</sup> It is added to the active material, conductive carbon, and binder material. When carrying out a solvent-based slurry, a preliminary dry mixing step is used to ensure a homogeneous mixture of the powders, including cathode active material (CAM), SE, and possibly conductive carbon, if not added to the binder solution.<sup>[17]</sup> Subsequently, the premixed solution of binder and solvent is dispersed with the dry mixture. Only the dry mixing step is required when creating a solvent-free slurry for dry coating, but different binders should be considered.<sup>[18]</sup> The

choice of the mixing recipe and procedure raises other challenges during mixing compared to LIBs, which will be further discussed in the following.

Significant changes producing the composite cathode arise due to the addition of SEs for SSBs compared to conventional LIB cathodes. The SE powder can vary in quality, depending on the synthesis method and supplier due to the currently low production volumes and novel synthesis routes.<sup>[19]</sup> The dry mixing step is decisive for a homogeneous distribution of various particle shapes and sizes. As shown in Figure 3a), long dry mixing times are necessary to break up agglomerates that are still noticeable for mixing times from one to five minutes. While, e.g.,  $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$  (NMC 622) or  $\text{LiNi}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1}\text{O}_2$  (NMC 811) active material typically has a  $d_{50}$  of around 2–10  $\mu\text{m}$  with spherical-shaped particles, the SE, e.g.  $\text{Li}_6\text{PS}_5\text{Cl}$  (LPSCl) or  $\text{Li}_7\text{P}_3\text{S}_{11}$  (LPS), is available in sizes ranging from below 1–8  $\mu\text{m}$  with a non-defined shape. Conductive carbon particles have a much smaller size down to the nanometer scale. Paring the suitable particle sizes of each material without causing agglomeration in the slurry poses a significant challenge from a production point of view. These aspects also affect the possible ratios of active material to SEs. Increasing the active material share inside the composite cathode leads to a higher energy density. Still, lithium-ions require transport paths along the SE particles as no liquid electrolyte is added.

The SE slurry production is less complex than the composite cathode slurry since only two components are needed. However, the SE limits the choice of binder and solvent.<sup>[19]</sup> Special attention must be paid to the evaporation rate of the chosen solvent. The widely used solvent toluene poses major concerns due to its rapid evaporation at room temperature and toxicity. The solvent choice influences the film's microstructure during drying (cf. Drying section), the production times, and the storage of the slurry.<sup>[20,21]</sup> Toluene-based slurries must always be sealed in a saturated atmosphere before the immediate coating. Otherwise, the fast evaporation leads to drying out inside of containers, tubes, or other stationary production points. This can also lead to longer cleaning times and more frequent maintenance intervals.

Comparing prepared slurries from literature and several experiments, the achievable solid content for the composite cathode in sulfide-based SSBs tends to be the same or lower as in LIBs. Usually, LIB slurries have a solid content range of about 60–80 wt%.<sup>[22–24]</sup> Having a composite cathode with sulfidic SEs leads to viscous pastes already at 60 wt% (active material and SE), but it is dependent on the choice of electrolyte and mixing technology.<sup>[17–20]</sup> The high solvent content has a negative impact on safety, costs, and poses environmental issues. Moreover, the SE slurry for the separator exhibits even lower solid contents between 30–60 wt%,<sup>[19]</sup> again depending on the powder properties and mixing technology. In summary, the



**Figure 3.** a) SEM images of composite cathode (CC) powder after 1–15 min dry mixing, b) Viscosity measurements comparing CC and solid electrolyte (SE) batches of 40 g mixed in a planetary mixer or a disperser (BC: binder content; SC: solid content), c) Photo of composite cathode slurry on the roll-to-roll coater at *iwb* (left: doctor blade side, right: exit after drying)

total amount of solvents in the composite cathode and SE slurry is high, leading to a major effort in planning and regulating the production periphery of the coating machinery and increasing energy consumption.

In LIB production lines, planetary mixers and dispersers are state-of-the-art for large-scale amounts of slurry.<sup>[25]</sup> These technologies can be used for sulfide-based SSBs as well. During the process development for both composite cathode and SE, the energy impact due to the rotational speed of either the mixing container (multi-axial system; speed mixer), a blade (disperser), or other tools must be scaled with the amount of material per batch. As presented in Figure 3b), the rheology of the produced slurry needs to be investigated due to the influence of the mixing tool, batch size, and further process parameters. Since already small amounts of slurry require a high energy impact to avoid agglomerates, scaling this parameter is critical when increasing the batch size.

Most important for sulfidic materials is the production under a very dry (dewpoints between  $-40^{\circ}\text{C}$  to  $-50^{\circ}\text{C}$ ) or inert atmosphere.<sup>[26,27]</sup> Available industrial mixers are usually equipped with an inert atmosphere or run under a vacuum, but adjustments regarding the material feeding and periphery must be considered. Preparation and weighing also have to be performed under an inert or dry room atmosphere.<sup>[27]</sup> Every transport container must be sealed and stable against corrosion.

## Dry Coating

The dry coating process is currently a widespread target for more sustainable and efficient production in LIB Gigafactories due to the absence of solvents and the shortening of the LIB production chain.<sup>[28]</sup> However, this is a complex process since new analysis methods are needed to characterize the powder mixtures, optimize the process, and, therefore, ensure the same homogeneous distribution of materials as in a slurry-based process.<sup>[29]</sup>

Concerning SSBs, adding SEs to the cathode mixture poses a significant challenge, especially considering the need for profound analysis tools to investigate the powder quality. Different dry coating approaches have been developed, such as dry spread deposition and hot melting extrusion.<sup>[29]</sup> Currently, these approaches show a low level of research. Dry electrode coating via dry-impact blending has been tested at a lab scale for sulfide-based half-cells.<sup>[30]</sup> However, the difficulties in implementing dry coating in LIB production show the numerous challenges of this technology, making wet electrode processing the most viable solution for advancing SSBs in the near future.

## Wet Coating

The wet coating process is the standard coating technique for battery production with advantages in terms of technology maturity. Slot dies are used as application tools for high coating

velocities, delivering precise mass loadings over wide coating widths for LIBs.<sup>[31]</sup> In a prototype or lab scale, the doctor blade is more accessible to implement and handle, significantly reducing the required minimal material amount compared to slot die coating and reducing the cleanup after production.<sup>[32]</sup>

Since the mixed slurry is the input material for wet coating, its rheology is the most influential parameter to align the coating process with the slurry behavior.<sup>[22–24]</sup> The need for more solvent in sulfide-based slurries compared to LIBs does not necessarily lead to lower viscosities due to the addition of SE to the cathode and differing particle sizes. Therefore, adapting the sulfide-based slurries to the viscosities known from LIB production is possible. However, in first studies, the absence of LIB typical plateaus of the dynamic viscosity at higher shear rates was observed.<sup>[19]</sup> These are usually significant properties exhibiting constant viscosities at low and high shear rates. Consequently, a different slurry behavior for sulfidic materials can lead to unforeseen challenges during coating, especially for high shear rates evoked in slot dies and necessary for high throughput production. A strong influence of the SE particle size and share on the viscosity was shown by Singer et al. (2023).<sup>[19]</sup>

The homogeneity of the coatings is essential since the SSB cell relies on solid-to-solid contact between components to provide sufficient ionic conductivity and low resistances at the interfaces.<sup>[33]</sup> Therefore, the roughness of coatings can be decisive to gain sufficient interfacial contact between electrodes and the separator. The sulfide-based SE is typically softer than the CAM, which is essential for the densification process and to ensure good contact with the electrodes. However, CAM and anode material are still deciding factors for the solid-to-solid contact at the interfaces.<sup>[34]</sup>

The atmosphere in the coating machinery must be dry air or inert gas. Depending on the requirements, mini- or micro-environments (cf. Figure 3c) are necessary to ensure the occupational safety of workers.<sup>[35]</sup> Essential changes compared to LIB production include the design of the application tool (slot die, doctor blade, etc.) in close alignment with the rheological properties of the sulfide-based slurry at high shear rates. Furthermore, the machinery and layout have to be adjusted to the respective coating strategy (cf. Figure 2).

## Drying

The drying process is the most energy-intensive process in a slurry-based route, with a significant impact on the homogeneous distribution of particles inside the coating.<sup>[36]</sup> In industry, either a convection dryer, an infrared dryer, or a combination of both is used to evaporate the solvent in a controlled manner.<sup>[37]</sup>

Significant changes arise in the drying process of sulfide-based SSBs compared to LIBs due to the previously mentioned increased solid content of the slurry (30–60 wt%) and the inability to use NMP or water as solvents for the composite cathode and separator. Choosing solvents with a higher evaporation rate, such as toluene, will decrease the drying times, leading to shorter dryer lengths.<sup>[21]</sup> However, the impact

on the microstructure might be significant. A very fast evaporation can lead to the migration of particles brought to the surface due to capillary forces.<sup>[20]</sup> Therefore, a thorough investigation of the cause-effect relationship between the solvent choice and drying process must be conducted.

The binder is typically the most temperature-sensitive material, limiting the maximum drying temperature and rate. Moreover, the properties of the binder, e.g., the density, the morphology, etc., influence the severity of binder migration.<sup>[38]</sup> To date, most of the studies about the LIB drying process and binder migration published have focused solely on polyvinylidene fluoride (PVDF).<sup>[39]</sup> New binder systems in SSBs need to be investigated in detail regarding their behavior during the drying process. A recently discovered phenomenon is the possibility of SE particle migration.<sup>[20]</sup> Since the particle size distribution of the SE is usually much smaller than for the CAM and the density is about a third of the CAM particles, the SE distribution is likely to change during drying. Also, the sedimentation of the CAM is likely to happen. Studies about drying in LIB production have shown that the drying rate majorly influences the adhesion strength.<sup>[40]</sup> First studies on similar correlations of the presence of binder migration and the decrease in adhesion strength were conducted by Singer et al. (2023)<sup>[20]</sup> for sulfide-based composite cathodes and SE separators.

Concerning the coating strategies described in 2a), a wet coating process for the SE slurry onto the composite cathode would require the drying of a multi-component system, as the slurry would partially penetrate the cathode substrate. In this case, a second drying of the multi-component material and the compatibility of the solvent from the SE slurry on an already-dried (and densified) cathode must be investigated. In summary, the use of drying equipment from LIB production in ASSB production does not require complex adjustments to the technology, despite the changed atmosphere.

## Densification

The state-of-the-art compaction process currently used for LIB electrodes is the calendering technology, where two rolls compress the component in between to the desired porosity by the applied line load. While LIB electrodes require densification to a defined residual porosity, the composite cathode and the separator in SSBs should theoretically have zero percent porosity. This is not achievable in praxis, but the lower the porosity, the better the SSB properties.<sup>[41]</sup> Higher porosities in SSB components were proven to lead to lower ionic conductivities, and lower total energy density of the cell because of residual pore volume.<sup>[42]</sup> As the SE particles have to facilitate the rearrangement of the hard CAM particles during densification, the SE share is a highly influencing factor for the densification process of composite cathodes.

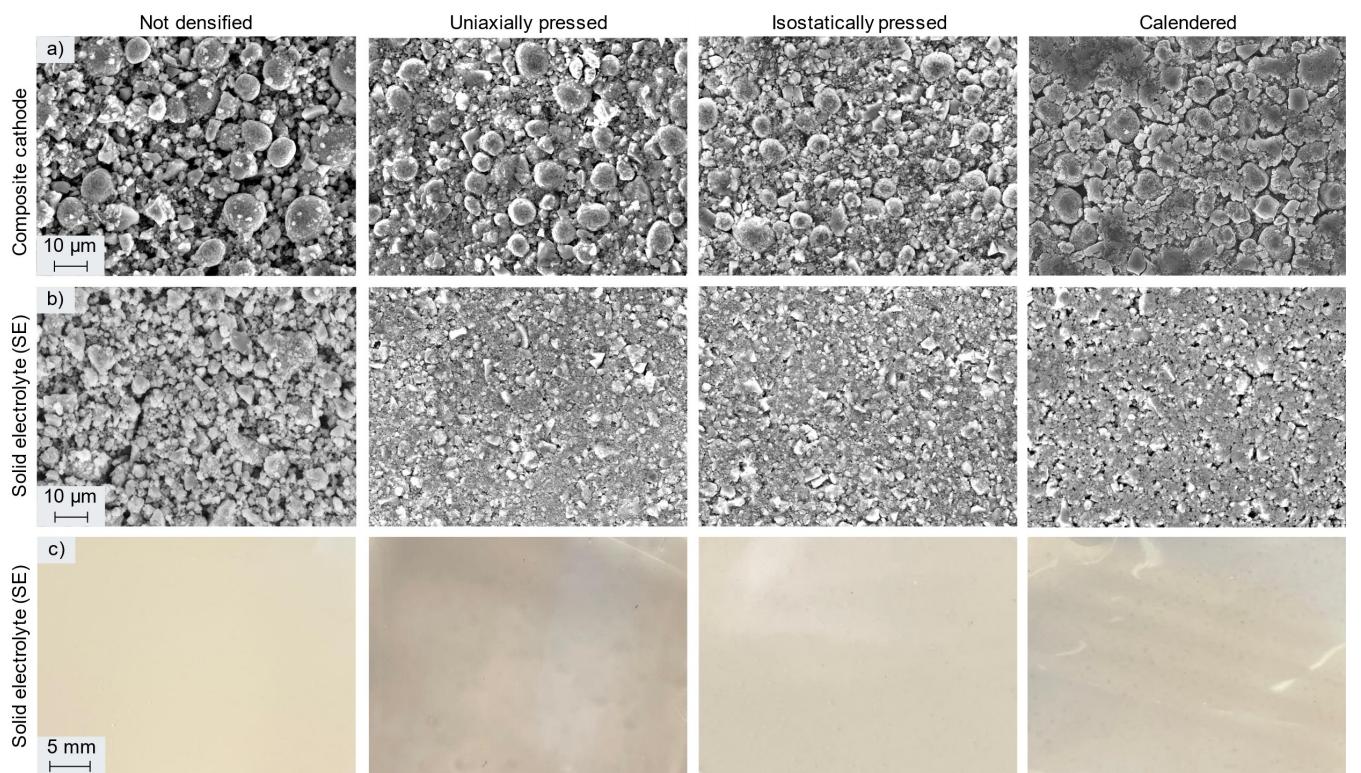
Reports show that immense pressures of up to 700 MPa at room temperature are needed to compact the material using a uniaxial press at lab scale.<sup>[42]</sup> The micromechanical processes occurring during the densification of the multi-material system of the composite cathode are complex and currently not well

understood. Simply increasing the compression forces to reach lower porosities is not feasible, as cracking of CAM particles can occur. CAM for sulfide-based SSBs is usually coated with a thin protection layer that inhibits the electrochemical reaction of the sulfide electrolyte with the CAM.<sup>[43]</sup> In case of cracking, this protective layer would break, leading to degradation and even failure of the cell in the long term due to the direct exposure of the CAM surface to sulfidic material. Moreover, the required high compression will lead to defects like wrinkling and corrugation of the materials,<sup>[44]</sup> especially in large-format components. Due to the significant material differences between LIBs and sulfide-based components, unknown challenges could arise. As the pressure in uniaxial pressing cannot be converted to a line load, densification studies must be conducted for the calendering process.

Considering the process chains pictured in Figure 2, different compaction scenarios can be drawn. A symmetrical coating on both sides of the substrate will be mandatory to decrease wrinkling due to unbalanced stresses during calendering.<sup>[44]</sup> While the densification of each individual component by itself poses a significant challenge, the densification of multi-components further increases complexity. However, multi-component densification would decrease the number of production steps in a scaled-up sulfide SSB production. The suitability of calendering for this densification task has yet to be reported systematically. However, an innovative technology, the isostatic pressing, is increasingly discussed in literature.<sup>[45]</sup> The advantage of isostatic pressing is the homogeneous force distribution on the samples surface, reducing unwanted deformation or defects. However, isostatic presses come with a large footprint and low throughput due to the discontinuous process and required times for pressure build-up and release. The pressure is applied to the sample by a surrounding liquid (oil or water)<sup>[58]</sup>, requiring the samples to be packaged. The industrial capability of this technology still has to be proven in terms of high-quality output and advantages compared to the widely established roll-to-roll technique calendering.

Figure 4 compares SEM images for sulfide-based composite cathodes in a non-densified state, and densified by uniaxial press, isostatic press, and calender. The final porosity of the respective samples needs to be investigated in detail. However, the microstructure exhibits differences in the roughness of the surface. Exemplary photos of the samples show that depending on the densification technology, the surface of the separator is influenced. Only the calendered separator shows a very smooth and shiny surface, while the uniaxial press equipment is darkening the color of the separator. Therefore, the choice of densification technology is significantly changing the morphology of sulfide-based components.

In summary, no published systematic studies on the densification of sulfidic components are available. Changes in process-product relations must be further investigated for each technology.



**Figure 4.** Row a) and b) SEM images of non-densified and densified SE separators (LPSCl) and composite cathodes (NMC 622, LPSCl) with uniaxial pressing, isostatic pressing and calendering; row c) Photos of respective SE separators with same composition and materials.

## Laminating

The lamination of separators with electrodes has been successfully applied in LIB production.<sup>[46,47]</sup> The main goal being the production of more robust intermediate products, removing the need to handle limp separator sheets while at the same time significantly improving the wetting behavior of the cell stack.<sup>[46]</sup> However, for sulfide-based SSBs, a lamination step can be crucial to enable their function.

The solid-to-solid contact of each component in sulfide-based SSBs leads to high interfacial ionic resistances within the cell, limiting the rate capability to a point where no actual use is possible. To counteract these boundaries, high operating pressures are required. Research has shown that improving the contact between SSB components during production can reduce the operating pressure needed for a LIB comparable cell performance.<sup>[48,42,14]</sup> A scalable implementation of this concept is proposed in the process chain pictured in Figure 2a). Here, the separator is coated on the cathode, maximizing the interfacial contact area. The densification of the separator after drying increases the contact at the interface and ensures a low porosity.

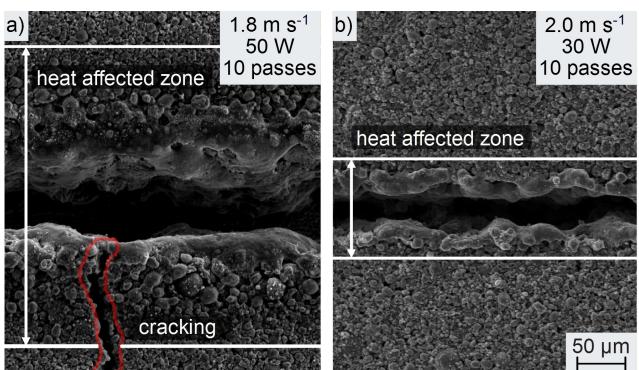
However, the separator-anode interface remains critical in the described process chain. A lamination step can be implemented in later stages of production to improve this aspect. Here, a pressure higher than the battery operating pressure is applied to laminate the anode onto the separator surface. This can be achieved continuously using a calendering

process or after the component cutting in a discontinuous process such as uniaxial or isostatic pressing. In addition, or alternatively, after the stacking process, the entire stack can be compacted, thereby laminating the components through uniaxial or isostatic pressing. Although the pressing of whole cell stacks is already part of some industrial LIB productions, new machinery is required for sulfide-based SSBs due to the necessary higher pressures. Depending on the necessary line load, existing LIB machinery can be reused to laminate continuous components, such as a silicon anode and the proposed cathode-separator multi-component.

## Cutting

Laser beam processing is one of the currently used technologies for cutting LIB electrodes due to fast processing times and low mechanical stress impact.<sup>[49]</sup> These qualities make the laser beam cutting process highly relevant for a scaled-up production of sulfide-based SSBs. However, systematic research on the application for sulfidic components has yet to be performed. Using an ultra-short pulsed laser process under an inert atmosphere, preliminary testing at *iwb* showed that laser cutting is feasible for a sulfide-based composite cathode, as shown in Figure 5.

As previously described, a cathode-separator composite can be advantageous in sulfide-based SSB production. To ensure functionality further down the process chain, the edges of this



**Figure 5.** SEM images of a laser cut in a sulfide-based composite cathode with an unoptimized parameter set a) and improved processing b); process-induced cracking and the heat-affected zone are marked.

composite must be in pristine condition, providing complete coverage of the separator on the cathode after the cutting process. Using mechanical punching could lead to smeared cut edges and delamination of the separator layer. Therefore, a precisely adjusted laser beam process is required to enable the defect-free cutting of sulfide-based SSB components.

The used materials and the thickness of the components in sulfide-based SSBs are similar to those in LIBs. Therefore, the laser beam systems employed in a sulfide-based SSB production will likely be comparable to the systems in a LIB production. However, there is currently no literature regarding the optical and thermal properties of the sulfide-based SE. Therefore, the material's behavior when submitted to the laser beam process cannot be predicted. Nevertheless, first experiments show positive results for the processability of a sulfide-based composite cathode (cf. Figure 5b). Further research is being conducted to optimize the laser beam parameters for processing the composite cathode and the SE, as well as multi-components. In contrast to LIB production, the laser beam cutting process in sulfide-based SSB production requires an inert environment, causing a need for new machinery and periphery.

## Stacking

LIBs can be assembled by z-folding, winding, lamination stacking, or single sheet stacking, with only the latter being widely used for lab-scale assembly of sulfide-based SSBs. Although z-folding has been proven feasible for sulfide-based SSBs,<sup>[7]</sup> single sheet stacking allows for higher energy densities on the cell level, making it an attractive technology for scaled-up SSBs.

Similar to LIB production, the anode is typically dimensionally oversized in sulfide-based SSBs to ensure a homogeneous current density on the cathode.<sup>[14]</sup> No systematical studies on the dimensional considerations for sulfide-based SSBs are currently published but could lead to a better understanding of the mechanisms at the electrode edges and be used for optimizing the cell design.

During the stacking of the SSB, the electrodes are ionically connected, enabling the cell electrically during the assembly process. Electrical isolation of the electrodes must be ensured to prevent a negative impact on later performance and safety issues, requiring preventative measures in the machine design process.

## Contacting

A serial (bipolar) contacting of elementary cells in a multi-layer sulfide-based SSB could enable higher energy densities on the battery and module level due to only requiring contacting of the outmost electrodes with an electrical tab. So far, multi-layer cells have mostly only been shown to work on a small scale with cells in the range of a few mAh<sup>[50]</sup> and might still prove impractical in an industrial application. As in LIB production, conventional parallel contacting of elementary cells is done in one or two subsequent steps, where the electrodes are electrically contacted, and a tab is welded onto them. The current state-of-the-art ultrasonic welding technology in LIB production is also suitable for sulfide-based SSBs. However, new materials for current collectors, especially in the context of bipolar cell configurations, might require the currently researched laser-based contacting due to its high flexibility and multi-material suitability.<sup>[51]</sup>

## Packaging

While prismatic, cylindrical hard case packaging, or pouch packaging are all currently used in commercial LIBs, only pouch packaging is currently suitable for sulfide-based SSBs due to their required high operational pressures. As both the cathode and the anode designs are not fixed at the current state of research, it is unknown if LIB pouch foils are suitable for multi-layer SSBs. E.g., a lithium metal anode poses a challenge to the pouch foil due to the significant change in thickness during cycling and the associated mechanical stresses, especially in multi-layer cells.

## Cell Finalizing

After physical production, LIBs undergo numerous formation cycles, where the cells are charged and discharged at low currents to form an interface between the liquid electrolyte and the active material.<sup>[11]</sup> It is not known if similar cell finalization is necessary for sulfide-based SSBs, as no studies have been published so far. However, it is assumed that no formation or only few cycles will be needed. In sulfide-based SSBs with silicon anodes, a higher volume expansion during the first cycles has been observed compared to the latter cycling, posing a challenge for holding the cell under constant pressure.<sup>[15]</sup> New cell testing equipment for SSBs is required as the high operational pressures cannot be applied with LIB cell testing equipment. More space must be allocated inside temperature

chambers typically used for cell testing and formation in industrial-scale production. The concept of holding multiple batteries in one pressure fixture during the formation and testing has been proposed.<sup>[7]</sup> Another aspect to consider during the formation and testing of sulfide-based SSBs is the monitoring of the changes in the thickness of a cell and the pressure, as these parameters could lead to valuable insight into the cell's electromechanical behavior similar to LIB cell finalization.<sup>[52]</sup>

## Production Environment

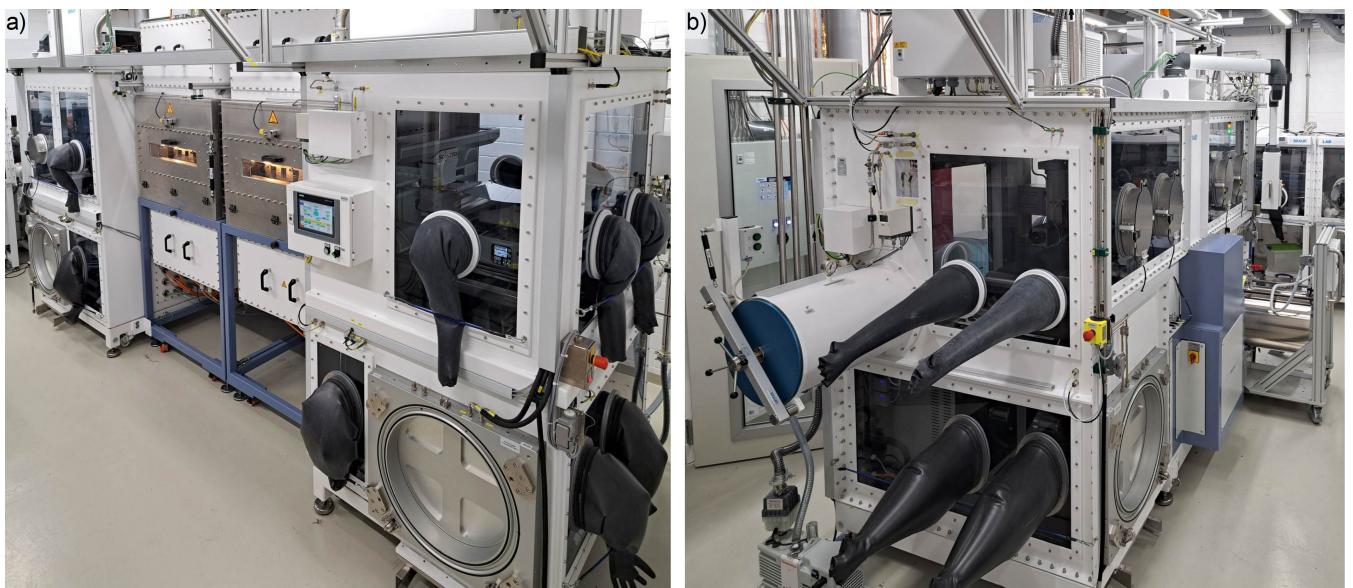
The design of the production environment is one of the most discussed aspects of sulfide-based SSBs on an industrial scale.<sup>[26,27]</sup> The hydrolysis of the sulfidic SE poses a significant challenge for the safety in SSB production. On the one hand, hydrolysis leads to irreversible chemical reactions and, therefore, decreases the quality in terms of ionic conductivity. On the other hand, the guarantee of occupational safety is of greater importance. Securing a constant and low dew point (less than -40 °C<sup>[27]</sup>) to impede the formation of large amounts of toxic hydrogen sulfide gas (H<sub>2</sub>S) implicates high costs and complexity, e.g. dehumidifying technology and safety concepts. Current state-of-the-art dry rooms achieve dew points down to -60 °C. However, a malfunction could lead to the accidental exposure of workers to high concentrations of H<sub>2</sub>S. Moreover, the human body radiates water through exhalation and through the skin. This water also reacts with the sulfide-based materials, leading to very concentrated H<sub>2</sub>S clouds in the near surroundings of the worker.

Separating the worker from the sulfidic materials would allow safer production and guarantee a high quality of the SE, even after manual handling steps. While on a laboratory scale, an inert atmosphere is easy to implement, e.g., in commercial gloveboxes, these systems are rarely used in high-throughput industries. The reasons are handling and maintenance issues and high prices for inert gases. High purchasing costs are caused by few available suppliers that integrate prototype or industrial battery machinery inside so-called microenvironments. The design of these machines is complex, as many electronic components are not designed for use in inert atmospheres due to the reduced heat dissipation and higher risk of flashovers. Moreover, the compatibility of materials in the machine with the sulfide-based material needs to be considered since, e.g., copper reacts with H<sub>2</sub>S and would, therefore, corrode or age faster than usual. Machinery with these requirements is not an "off the shelf" product but needs to be designed with high effort and precise knowledge about the demands coming from sulfidic materials. Besides the manufacturing processes, production involves further and mostly non-value-adding aspects such as material storage, transportation, etc. Considering the sensitivity of sulfides, risks and safety assessments need to be rethought compared to LIB production throughout the factory.

To summarize, the key challenges in the production of sulfide-based ASSBs along the process steps are outlined in Table 1. The most important key factors are differentiated for material, process, and machinery level.

**Table 1.** Key challenges in transitioning from lithium-ion battery production to sulfide-based solid-state battery production on material, process, and machine levels given for each process step. The described challenges focus manufacturing in microenvironments under an inert atmosphere.

Production step	Material	Process	Machine design
Mixing	Matching particle size distribution of SE and CAM Overcoming initial agglomeration of SE Ensuring solvent compatibility	Controlling temperature during dry mixing Achieving process stability	Mixing in closed containers Preventing corrosion caused by aggressive solvents
Coating	Dealing with unknown rheological properties	Working with small viscosity windows Developing coating strategies	Handling additional auxiliary substrate foils Designing suitable coating tools
Drying	Removing increased solvent shares	Handling fast solvent evaporation rates Minimizing binder and electrolyte migration	Removing solvents from closed production atmospheres
Densifying	Avoiding CAM damage Achieving virtually zero percent porosity	Identifying suitable densification technologies	Achieving high densification pressures
Cutting	Dealing with multi-layer compounds	Ensuring high edge quality	Processing under an inert atmosphere Minimizing particle contamination in closed systems
Stacking	Handling fragile components	Handling electrically active (multi-) components	Full automation in an inert atmosphere
Welding	Adopting new current collector materials	Handling electrically active (multi-) components	Integrating machinery in an inert atmosphere
Packaging	Managing electrode expansion	Handling electrically active (multi-) components High operation pressures Cell breathing	Full automation in an inert atmosphere



**Figure 6.** Photos of the SPACe<sup>-</sup> (Sulfidic Cell Production Advancement Center) at *iwb* (at TUM); microenvironments for the roll-to-roll a) Coating and b) Calendering process of sulfide-based composite cathodes and SE separators.

### Academic Research at Industry-Relevant Scale

At the *iwb*, a pilot-scale production line for scalable and semi-automated manufacturing of SSBs is set up (cf. Figure 6). In the SPACe<sup>-</sup> (Sulfidic Cell Production Advancement Center), unique machinery housed entirely under an inert atmosphere for the production of sulfide-based SSBs is used for research. The machinery was designed and constructed to ensure the fulfillment of requirements for the fabrication of new materials. The connection between the coating and the calendering machine is realized through portable carts to insert or eject sulfide-based material coils. Moreover, short-term storage of coils or sheets under an inert atmosphere or vacuum is possible inside the carts. Cell assembly occurs in another glovebox system equipped with cutting, stacking, welding, and packaging machinery.

The scale of fabricated multi-layer SSB pouch cells is within the range of approx. 1–5 Ah, paving the way to the proof-of-concept for a roll-to-roll production validated with prototype cells.

Considering the TRL scale for EV battery application, the goal of the SPACe<sup>-</sup> is to bridge the gap between research laboratory and pilot line battery production, contributing to an increase of the sulfide-based SSB TRL to levels 5 and 6.<sup>[53]</sup>

### Conclusions

This study comprehensively overviews a sequence of processes to define a scalable sulfide-based SSB production. The introduction of the single- and multi-component levels is intended to provide a framework for the better comparison of differences between LIB and SSB production. A detailed description of the required changes in process parameters and machine design

compared to LIBs, based on insights into experimental results, is highly valuable from a production point of view. The summarized key challenges pave the way for future production research to accelerate the development of competitive SSB technology. The need for highly automated and aligned machinery to fabricate SSB components is evident. The SPACe<sup>-</sup>, an innovative production line at the *iwb* at the Technical University of Munich, provides a first step towards the technology's scale-up.

### Acknowledgements

The authors wish to thank the Bavarian Ministry of Economic Affairs, Regional Development and Energy for funding the research project "Industrialisierbarkeit von Festkörperelektrolyt-Zellen." They also wish to thank the German Federal Ministry for Economic Affairs and Energy and the Bavarian Ministry of Economic Affairs, Regional Development and Energy for funding the work in the BMW project "IPCEI EuBatIn" (16BZF205). The authors thank BMW for the cooperation and MBraun, Mathis, Scantech, and Quintus Technologies for their cooperation and supply with machinery within this project. A great thanks goes to Sophie Grabmann. Open Access funding enabled and organized by Projekt DEAL.

### Conflict of Interests

The authors declare no conflict of interest.

## Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

**Keywords:** Solid-state batteries · Sulfide-based electrolyte · Solid electrolyte · Production technology · Battery production

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Manuscript received: March 2, 2024

Revised manuscript received: April 23, 2024

Version of record online: May 27, 2024