

Scalable Processing Routes for the Production of All-Solid-State Batteries—Modeling Interdependencies of Product and Process

Célestine Singer,* Joscha Schnell,* and Gunther Reinhart

The all-solid-state battery (ASSB) based on a solid ionic conductor is a significant future concept for energy storage. In respect of the growing global demand for batteries, a systematic study on processing thin-layer and large-area ASSBs is addressed herein. As ASSB cells are mainly produced on a laboratory scale, an introduction to industrial production is needed. Therefore, the formation, ranking, and selection of technology chains are presented concerning the strategic orientation of cell manufacturing companies. A system model consisting of five sub-models is created, which connects technologies with production-relevant parameters. The results are used for a tool that automatically generates and evaluates technology chains in dependence of the ASSB cell design. Starting from the layer fabrication technologies further up- and downstream processes are defined. For sulfidic solid electrolytes, e.g., a ball milling followed by the aerosol deposition method, hot pressing, and laser cutting are favorable in terms of high-quality layers and low production volume, whereas planetary mixing, tape casting, calendaring, and die cutting are the choice for a high-throughput production. Based on these findings, processing routes for every cell design and solid electrolyte material can be generated concerning company-specific criteria, thus enabling the industrial application.

1. Introduction

The expansion of renewable energies and the enactment of laws to reduce emissions are encouraged by climate policies. As part of this, the electrification of the global automotive market is progressing but still suffers from customer unacceptance.^[1,2] In


the future, the usage of electric vehicles will highly depend on the progress of associated technical core components such as the lithium-ion battery (LIB).^[3] The development of energy storage, therefore, is of decisive importance to optimize sustainable energy systems and to mitigate environmental pollution.^[4,5]

LIBs are the key technology in electric vehicles to accomplish market and customer requirements.^[6] These relate especially to the driving range and the charging time^[7] throughout operational safety.^[8] The main proposed advantage of the all-solid-state batteries (ASSBs) is their increased safety, which results from replacing the flammable and toxic liquid electrolyte in LIBs with a solid ion conductor. Furthermore, the application of metal anodes, e.g., pure lithium, to enhance the energy and power density is discussed to be most likely achieved by the use of a dense and mechanically stable solid electrolyte.^[9] Although polymers, sulfidic, ceramic oxide-based and halide materials are identified as promising solid-state electrolytes,^[10] challenges arise in the identification of material compatibilities, suitable cell designs, and production technologies.^[11] Yet, the development of production processes for thin-layer^[12] and large-format ASSBs is crucial for successfully bridging the gap between laboratory research and the industrial market.^[13,14]

The production of ASSB cells for electrochemical research purposes and first process sequences of different technologies are described in various publications.^[11,15–24] However, a systematic study on processing routes considering every value-adding production step and related parameters in dependence of the cell design is omitted. Novel materials and components of ASSBs are characterized by interdependencies among each other,^[25] which in turn are affecting the respective production steps.^[19] (Electro-)chemical obstacles regarding the choice of materials, blocking interfaces, and instabilities influence the scalability of processes. The high uncertainty about a suitable production line, therefore, has a major impact on impeding the industrial application.^[21]

In this article, a detailed system model for ASSBs and scalable production technologies is presented. Interdependencies between the processes and the product structure of ASSBs are considered. The method consists of five sub-models, which

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are described exemplarily for a sulfidic ASSB cell. As a result, different process routes for the sulfidic cell are shown and discussed concerning their suitability for company-specific criteria. The results highlight, that processing routes for various cell designs can be defined in an early stage of technology readiness level^[26] for ASSBs. The consideration of multiple product and process parameters, however, is essential to establish processing routes and implement ASSB production in the industry.

2. Fundamentals

2.1. Solid Electrolyte Separator Materials and Production Challenges

The most promising solid electrolyte separator (SES) materials for electric vehicle applications are categorized as organic polymers^[27] or inorganic sulfides and oxides^[28,29] or halides.^[30] The focus of this study is mainly on sulfide materials as their high ion conductivity as key property enables them to be used in electric cars.^[31–33] Concerning processing of different SES materials, challenges arise indicating the complexity of the production system. Although the thermal and chemical properties of oxides are of great advantage for the application in ASSBs,^[34] drawbacks are evident due to their high interfacial^[35] and grain boundary resistances,^[36] which reduce the ionic conductivities.^[37] Therefore, a complex sintering step at high temperatures ($\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZ) $\approx 1100^\circ\text{C}$; $\text{Li}_{1+x}\text{Al}_x\text{Ti}_{2-x}(\text{PO}_4)_3$ (LATP) and $\text{Li}_{1+x}\text{Al}_x\text{Ge}_{2-x}(\text{PO}_4)_3$ (LAGP) $\approx 800^\circ\text{C}$.^[38] A reduced sintering temperature for LLZ can be achieved by adding additives, e.g., Al_2O_3 ^[39]), which implicates difficulties and high costs in scaling up, is required to densify the oxide layers and increase ion conductivity.^[40] If two or more cell layers are co-sintered, chemical reactions in between these layers may occur which damages, e.g., active material or binder.^[41] Furthermore, sintered oxides exhibit high stiffness and brittleness, which makes processing during ASSB production more difficult.^[42] Halide materials have shown a fast development over the last few years, but still suffer from poor reduction stability and their ionic conductivity needs to be improved.^[30] The advantage of sulfides is their high conductivity without the need for a sintering step.^[43] In most cases, mechanical pressing of these materials is sufficient to form a dense layer.^[44] Heat input is not necessary^[45] but can lead to higher conductivities.^[46] Accordingly, the grain boundary resistances are not as dominant as in oxide materials.^[47] An obstacle of sulfides is their narrow electrochemical stability window and the instability of some sulfides against cathode active materials and lithium metal as anode.^[48] For the cathode side, an active material coated with additional layer of, e.g., LiNbO_3 , is commonly used to suppress unwanted side reactions.^[49] As few sulfide SES are stable against lithium metal, protective layer such as polymers or LiPON are used to stabilize the interface.^[35,50] The main disadvantage for the use of sulfidic materials in production is the instability toward water in an atmospheric environment, where H_2S , a toxic gas, is easily formed.^[51,52] For this reason, processing must take place in a dry room or inert gas atmosphere.^[53,54] Another major challenge remains the integration of lithium metal into the ASSB to further increase the energy density of the cells. In addition to the

materials, the cell design plays a decisive role in the use of the lithium metal.^[55] Grain boundaries and defects at interfaces, for example, promote the rapid formation of lithium dendrites. As a result, high demands on the quality of the layers and thus the challenges for production become apparent. Furthermore, the handling of lithium metal is further complicated by properties such as adhesiveness and the thin layers required ($\approx 20\ \mu\text{m}$).

2.2. ASSB Cell Design

Typically, two types of ASSBs are distinguished according to their cell design. First, there are thin-film batteries which have already been established in series production for some applications.^[56] However, the limited electrode thickness (usually less than $10\ \mu\text{m}$) does not allow high energy densities, which are required in electric vehicles. In contrast, bulk-type batteries with a high layer thickness have been manufactured mainly on laboratory scale.^[57] Schnell et al.^[19] present two bulk-type battery cell concepts which are designed for a large-scale serial production. One of them is the cathode-supported cell, in which the cathode composite (Composed of active material, binder, carbon black, and solid electrolyte. A coating layer on the cathode surface can be necessary to obtain interface stability to the sulfidic electrolyte^[58]) is coated with a thin electrolyte layer.^[59] The anode either is applied in form of a lithium foil or deposited by, e.g., physical vapor deposition (PVD) or (in situ) electrochemical plating. The other concept is based on a three-layer matrix structure (“Tri-layer”) made of a dense SES located between two porous solid electrolyte layers.^[60] The cathode and anode materials are infiltrated into these porous structures. This approach originates from the production of solid oxide fuel cells (SOFCs) and is potentially suitable for the production of oxide ASSBs.^[61] Both cell designs need to be considered while developing concepts for the manufacturing process. Depending on the choice of SES material, an additional protective layer, e.g., made of aluminum oxide (Al_2O_3) or a (ion conducting) polymer, between anode and SES might be necessary.^[62]

2.3. State-of-the-Art Manufacturing Technologies for ASSBs

In addition to the identification and research of new SES materials, the development of manufacturing processes plays a decisive role.^[43] In previous publications, ASSBs have mainly been considered in the form of pressed pellets.^[63] This method is common for sulfides, as cold pressing leads to sufficiently low grain boundary resistances^[64] which results in a sufficient ion conductivity. However, in comparison to sheet-type batteries as shown by Yamamoto et al.^[37] and Kasemchainan et al.,^[65] the production route for pellets is not suitable for industrial scale.^[19,66] Currently, there are many technologies which are potentially usable to process different SES materials on laboratory scale.^[67] In addition to wet chemical coating processes (e.g., tape casting or screen printing),^[56,57,68] infiltration of SES solutions,^[69] gas phase depositions (chemical vapor deposition [CVD],^[70,71] PVD, pulsed laser deposition (PLD),^[72–75] and aerosol deposition method (ADM)^[76,77]) have been applied. In

addition, first publications connect different technologies to generate a process chain. For example, Baek et al.^[78] documented various layer fabrication technologies with regard to the necessity of a subsequent sintering step. However, in the publication of Troy et al.,^[16] environmental influences resulting from the manufacturing process of an ASSB were investigated. In Hu,^[15] a powder pressing process, a pasting route from SOFCs and a wet coating process are introduced as possible strategies to fabricate ASSBs. Most recent publications by Kerman et al.^[17] and Schnell et al.^[19,21] provide a detailed compilation of different technologies and suitable technology chains for the manufacturing of ASSBs. A latest review of Famprikis et al.^[66] shows an overview on currently available methods for processing solid electrolytes with a dry or wet route.

All of these approaches do address different possibilities to fabricate single sheets in an ASSB, but they do not correspond to the complexity of an ASSB resulting from uncertainties in material selection and the cell design. Combining production technologies to chains as well as a multi-criteria recommendation for a series production is not examined dependent on variations of the cell structure. To meet these requirements, a systematic and hierarchical structure of the ASSB as product

is needed. To enable the production of the complex ASSB cell designs, the identification and characterization of suitable technologies is addressed. By connecting technologies with production-relevant parameters, e.g., process time, quality, or maturity, the individual corporate strategy is considered to guarantee the entrepreneurial and high volume orientation of the processes.

3. Procedure for System Modeling

3.1. Outline of the System Model

By developing a system model (Figure 1), all occurring product-process interactions and the complexity of different cell concepts are addressed. The system model (Due to the interdisciplinary approach of Systems Engineering^[79] and the model-based system development,^[80] all system models were created with the graphical modeling language SysML^[81]) consists of three blocks: system requirements, system structure, and system behavior.^[82] The arrows show the linking between sub-models of the system structure and behavior, which serve as in- and output for other sub-models.

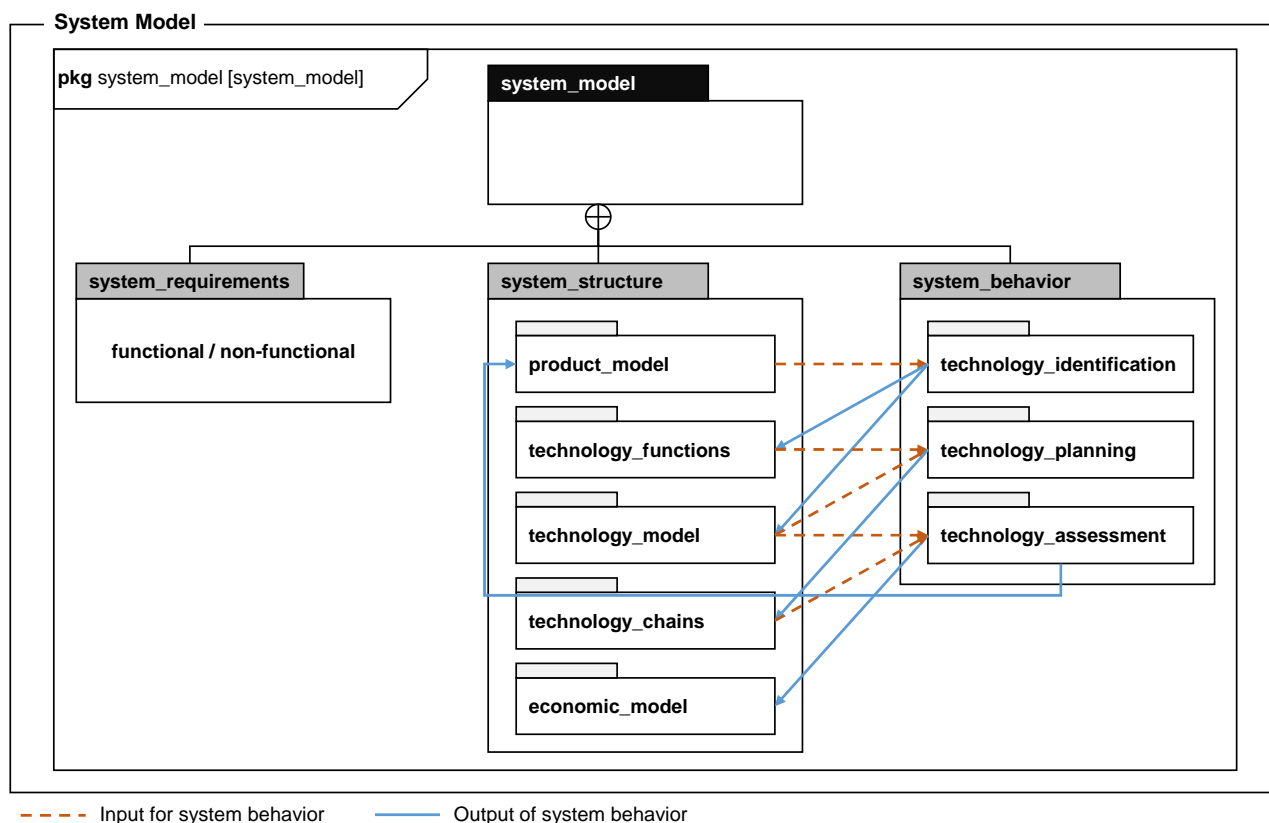


Figure 1. Overview of the System Model and the linking between sub-models of the system structure and behavior. Functional requirements address the dynamic behavior of the system and are defined as following: orientation on elementary aspects of the strategic technology planning, consideration of the status of development and complexity of the product, consideration of the status of development of the technologies, integration of the companies' strategy. The nonfunctional requirements address the quality standards of the system and, therefore, are not decisive for the functions of the system. For this model, following nonfunctional requirements were chosen: modularity of the models and methods, scalability of the system model, validity of the models and methods. For further explanation of the input and output parameters, please refer to the main text.

3.2. Structure

The system structure contains the product model to describe the ASSB components. The technology functions^[83] are derived from the product and consist of substantive-verb compositions to describe the production task, e.g., layer coating. Furthermore, the technology model is part of the structure to identify single processes. To define the entire technology chains, processes are connected considering their compatibility with the cell design and dependencies among each other. An additional economic model (Figure S11, Supporting Information) serves as an estimation of production costs.^[22] The detailed models are shown in the Supporting Information.

3.3. Behavior

The system behavior describes the interactions between the structural models and is subdivided into technology identification, planning, and assessment. The technology identification is performed according to Greitemann^[84] to find processes which are able to produce the single ASSB components. This identification is linked to the product as well as technology function model. The technology planning contains the generation of technology chains. Here, chains for the single components of the ASSB are defined and their compatibility is proven before linking them to a complete chain. The technology assessment is performed in an automatized tool, which compares technology

chains with each other and ranks them by considering corporate strategic parameters in dependence of the ASSB cell design. As strategic parameters, companies can specify their planned production volume as well as their prioritization of either technology maturity, product quality, or throughput. The relationship between input and output of every model is examined and defined as an interaction according to technology planning. Interactions can occur in between ASSB product elements, technology functions, and technologies.^[85] The system structure and the system behavior cannot be considered separately as they influence each other. Therefore, models of the system structure serve as a database for the behavioral aspects, which build the frame of this work.

4. Technology Identification

The product-technology-assignment (Figure 2) is the first behavioral procedure of the system to identify suitable technologies. The technology functions serve as basis for the identification of technologies.^[84] The technology model serves as database for the evaluation of the technical feasibility. Based on an ASSB product model (Figure S1 and S3, Supporting Information), technology functions are identified by deriving them from single product components.^[84] In a next step, exclusion criteria, e.g., error rate, are defined to evaluate the technical feasibility of technologies. As suggested by Fallböhrer,^[86] categories for criteria are material, product, and production requirements.

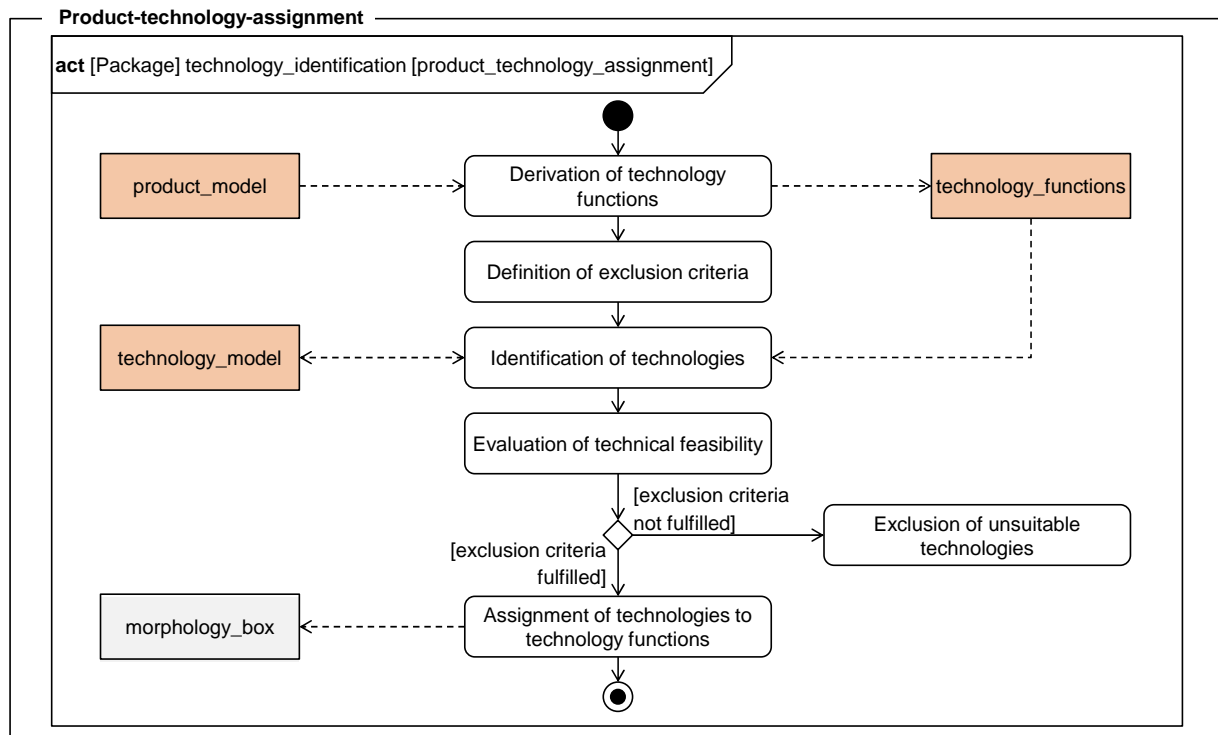


Figure 2. Activity diagram for the product–technology–assignment. Colored blocks are sub-models, which function as input or output. The diagram starts with the derivation of technology function and leads to the definition of exclusion criteria as well as the identification of technologies. The fulfillment of criteria is based on expert interviews presented in ref. [21]. The assignment of technologies to technology functions can be shown in a morphology box, which acts as an overview on possible technology chains.

Requirements from the product are then compared and suitable technologies are chosen. Technologies not meeting the exclusion criteria are dismissed, whereas all potentially suitable technologies are assigned to the respective technology functions and further described by detailed properties in a technical aptitude.

The realization of the product–technology–assignment is exemplarily carried out on basis of the sulfidic ASSB. As a first step, the technology functions, e.g., layer fabrication, for the ASSB production were derived from a product model consisting of a detailed ASSB and galvanic cell model (Figure S3, Supporting Information). The identification of exclusion criteria and evaluation of technical feasibility were conducted following the methods presented in ref. [21]. Requirements in terms of thermal stability of coatings or substrates (material criteria), throughput or production environment (production criteria) and layer thickness, geometry or error rate (product criteria) of each technology were analyzed. As a result, the technologies tape casting, screen printing, roller coating, spray coating, ADM, inkjet printing and PLD, which were allocated to the function “layer fabrication”, can be applied for the cathode composite as well as for the SES in a sulfidic ASSB (Figure 3). As a result of the expert assessment of the technology extrusion, this technique can only be used to fabricate the cathode composite,^[87] because its error rate is too high for applying it for the fabrication of SES layers. The reason are difficulties in producing extremely thin SES layers by extrusion.^[88] Screen printing and

electrophoretic deposition (ED) were identified to form only the sulfidic SES, as an adequate thick layer for cathodes are not possible to produce in a short time. The compaction of the cathode or SES can be achieved by a calender or uniaxial pressing; a punching or laser cutting process is suitable for cutting the components into shape. The structural models of the technology identification are detailed in the following.

4.1. Product Model

The product model serves as input for the product–technology–assignment and is divided into an ASSB model (Figure S1, Supporting Information) on product level as well as a galvanic cell model (Figure S3, Supporting Information) on module level. The ASSB can be described by several properties and includes the cell stack consisting of multiple galvanic cells and the cell casing. The cell stack properties are governed by the stacking sequence.^[20]

The galvanic cell (The galvanic cells can be stacked in parallel, with one of each anode and cathode layers sharing one current collector as a bipolar stack where each current collector is sandwiched between one anode and one cathode layer (Figure S2, Supporting Information)^[20]) of an ASSB is composed of one or two current collectors, the anode layer, a SES layer, and cathode layer (Figure 4). The collector foils are defined by their material properties, layer thicknesses, and the cost per unit area

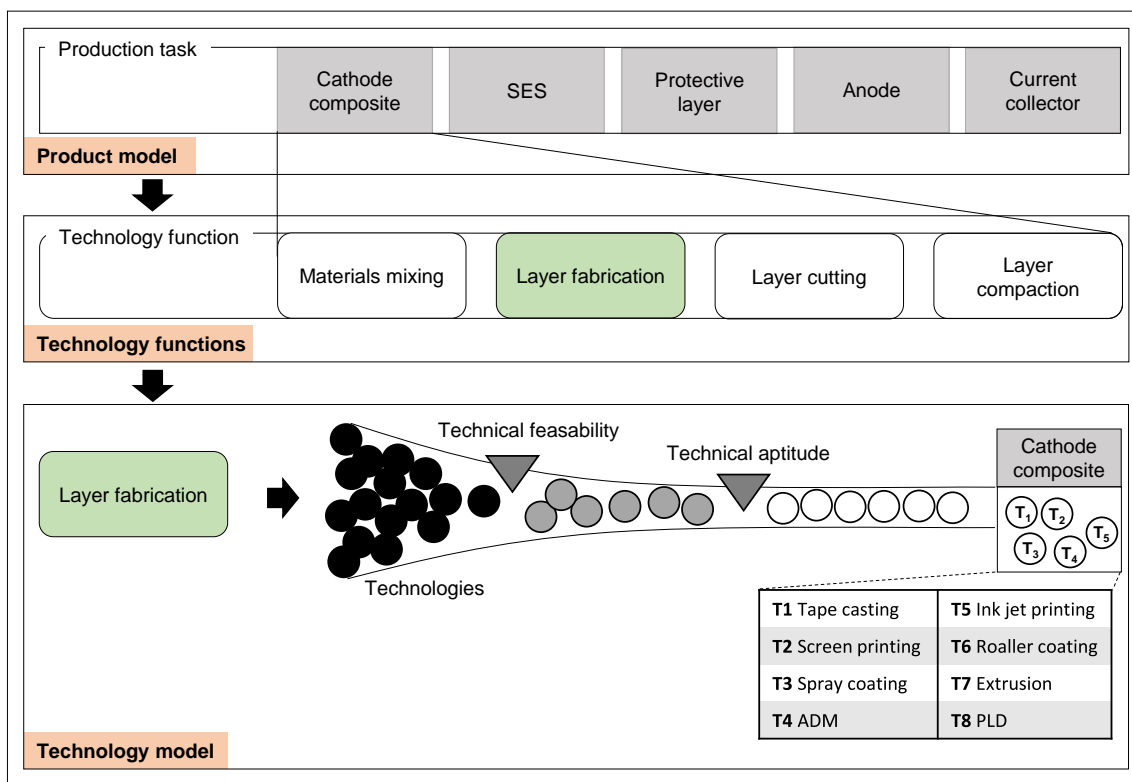


Figure 3. Schematic diagram for assignment of technologies to ASSB components consisting of product model, technology functions, and technology model with resulting technologies (T1–T8) identified for sulfidic layer fabrication. Exclusion criteria are assessed in the technical feasibility study to exclude unsuitable technologies. The technical aptitude is used to characterize the technologies and deposit values for selected properties.

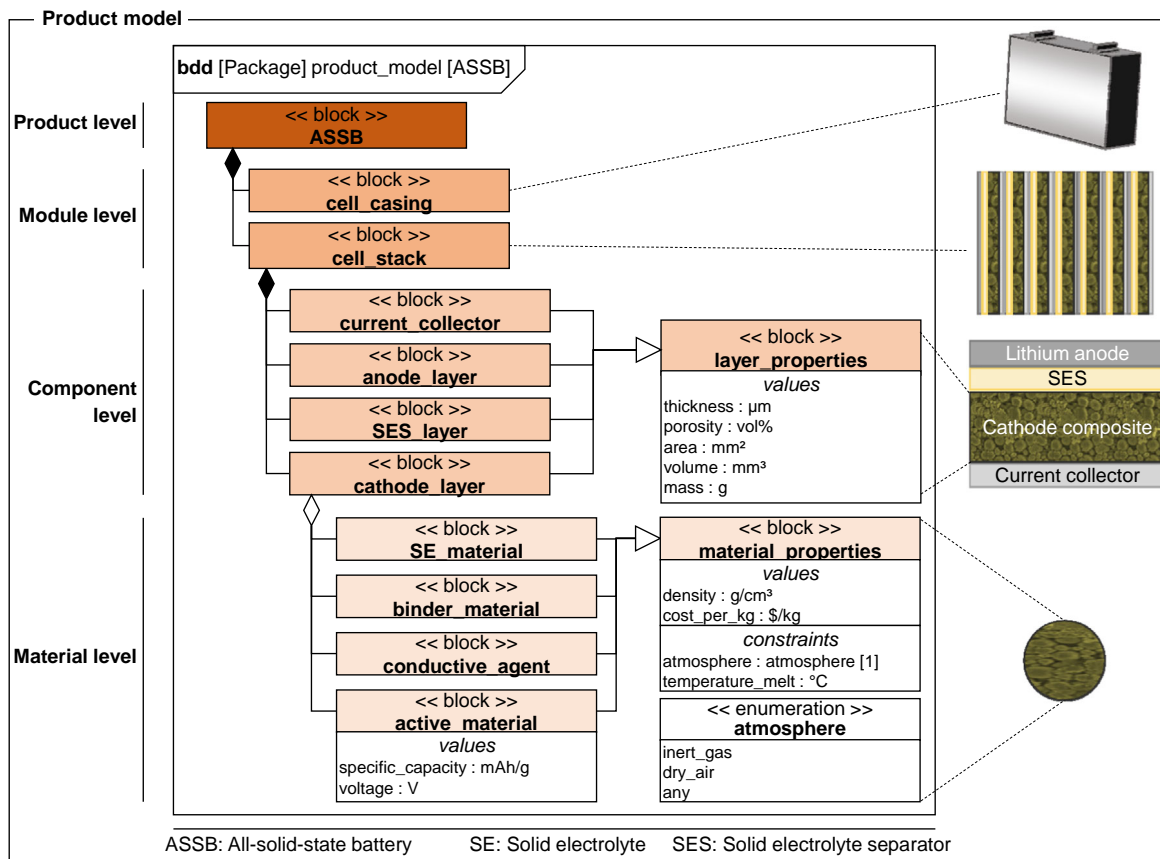


Figure 4. Excerpt of the block definition diagram for the product model of an ASSB. The properties of the ASSB are inherited to all subordinate components. Given values are exemplarily chosen and can be extended.

(in $\text{\$ m}^{-2}$). The thickness of the cathode layer and SES layer are used as input parameters. The anode layer thickness (T_{anode}) is calculated based on the area specific capacity of the cathode (C_{cathode}), the specific capacity of lithium metal (C_{LiM}), the density of lithium metal (ρ_{LiM}), and the area of the anode (A_{anode}), considering a balancing factor (BF_{anode}) (see Equation (1)).

$$T_{\text{anode}} = \frac{(C_{\text{cathode}} \times BF_{\text{anode}}) / C_{\text{LiM}}}{\rho_{\text{LiM}} A_{\text{anode}}} \quad (1)$$

The cathode layer is typically composed of active material, conductive agent, binder material and SE material, whereas the SES layer comprises the SE material and a binder.

Multiple SES layers can be attributed to one cell design to account for additional protective layers. All layers can be further described by layer properties, such as porosity, area, volume, and mass. The specific capacity and voltage are important attributes of the active materials. General material properties are the crystallographic density (in g cm^{-3}) and cost (in $\text{\$ kg}^{-1}$), which are important input parameters for calculations regarding the ASSB properties. Furthermore, the atmosphere of the production environment needs to be considered within material properties (Figure S3, Supporting Information).

4.2. Technology Model

As a result of the technology identification, the technology model can be constructed by organizing processes derived from technology functions shown in Figure S4, Supporting Information. Technologies were summarized in a technology pool to systematically categorize them with their abilities and constraints, based on the empirical study in Schnell et al.^[21] Value-adding production technologies (Figure S5, Supporting Information) were further classified according to DIN8580,^[89] as for instance shown in the example for coating technologies. A further specification takes place based on the input states, such as liquid/paste, powder, gaseous, or ionized.

Identified technologies, which are able to fulfill the requirements of the cathode composite layer fabrication in a sulfidic cell, comprise tape casting, screen printing, spray coating, roller coating, inkjet printing and extrusion (Figure S5, Supporting Information). Those processes can be assigned to coating technologies with a liquid/paste input material characteristic, whereas ADM uses powder as starting material and PLD is based on a gaseous material state. A production technology refers to the associated technology function and to the product components as shown by the morphology box (Figure 5). The core technology of this chain is associated to the layer fabrication. Supporting

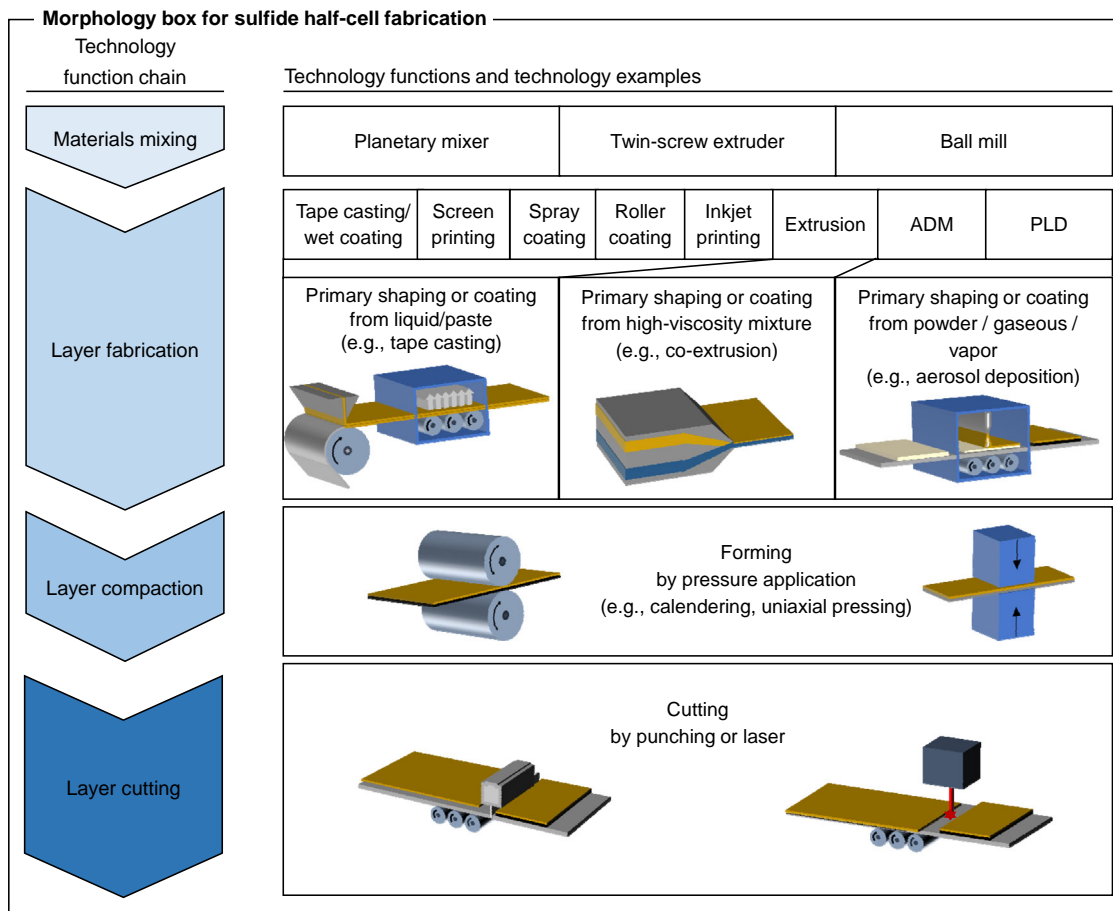


Figure 5. Morphology box on sulfidic cell fabrication for cathode composite and SES (half-cell) showing the assignment of technologies to technology functions.

technologies for the sulfidic half cell (cathode composite and SES) were defined per technology function. Materials mixing can be executed by planetary mixing, twin-screw extrusion, or ball milling. Calendaring or uniaxial pressing are preferentially used for layer compaction. Die or laser cutting is used for separating layers. At this point, several technology chains can be combined with these technologies (Figure 5).

5. Generation of Chains

To generate a technology chain, the fabrication of the individual components needs to be regarded (Figure S7, Supporting Information). A core technology and supporting technologies are selected from the morphology box. At first, the output of the core technology is compared with the desired end-product state of the component defined by the superordinate technology function. If all states match, the technology is suitable to produce the component without the need for supporting technologies. For the selection of suitable supporting technologies, interactions with other technologies in the component chain must be considered (technology–technology interactions). Therefore, intermediate states, which show differences in product states

are compared. Here, characteristics such as the porosity, the geometry, and the supply with single sheets or rolls are of great importance. If all intermediate states match, the final component chain is selected.

The comparison of the core technology output with the component features is demonstrated using the example of tape casting (wet coating), which is one of the most promising techniques to fabricate the cathode composite continuously. The substrate carrying the slurry containing the coating material is fed as rolled goods into the tape-casting process to maintain continuous processing. If a continuous process is desired, the binder content of the cathode should be adjusted to the roll-to-roll process. The thickness of the coated layer was set to 70–150 μm ^[21] (in dependency the energy density of the cell). These two restrictions are met by tape casting. The final state also includes a cathode layer with a defined cell format. The produced layer is processed as rolled material. As the required porosity of the sheets may not be reached after the coating process,^[23] the deficits of the technology tape casting, thus, amount to an insufficient porosity of the layer as well as missing cell sheet format. The deficits of the technology were compensated by selecting suitable supporting technologies (Figure 5), which were characterized in the technology properties model (Figure S6,

Supporting Information). Here, calendring can be used to compact the cathode composite and, thus, reduce its initial porosity. Punching or laser cutting can be applied to separate the layers, as those technologies were identified before. To determine the sequence of the technologies in the chain, the input and output states (intermediate states) of the supporting technologies are considered. Intermediate states show differences in the component features. For the cathode material, the calendring process needs a continuous material supply as input. Therefore, the technology laser cutting (or punching) must be used after compaction as it has a continuous material supply as input and an intermittent material supply as output. The required state of the component is fully achieved by both technologies.

A promising alternative is the extrusion as core technology, which enables to combine the mixing step with the coating step.

Here, also a solvent-free process is conceivable. Similar to the tape-casting process, the extrusion does not meet the restrictions of the cell format as well as the porosity. Therefore, after the extrusion a calendring step and separation by punching or laser cutting will be necessary.

The procedure shown in **Figure 6** was conducted for all cell components leading to the complete component chain model. In the next step, the technology chain is generated by combining the individual chains for the fabrication of single components (Figure S8, Supporting Information). Again, the core technology of each component chain is selected to investigate if a substrate is required for layer fabrication. If the component has to be coated or joined with another component or the intermediate state of a component, the interactions of all technologies and technology functions in the downstream must be considered. Hence, unsuitable component chains are excluded at this stage. If all

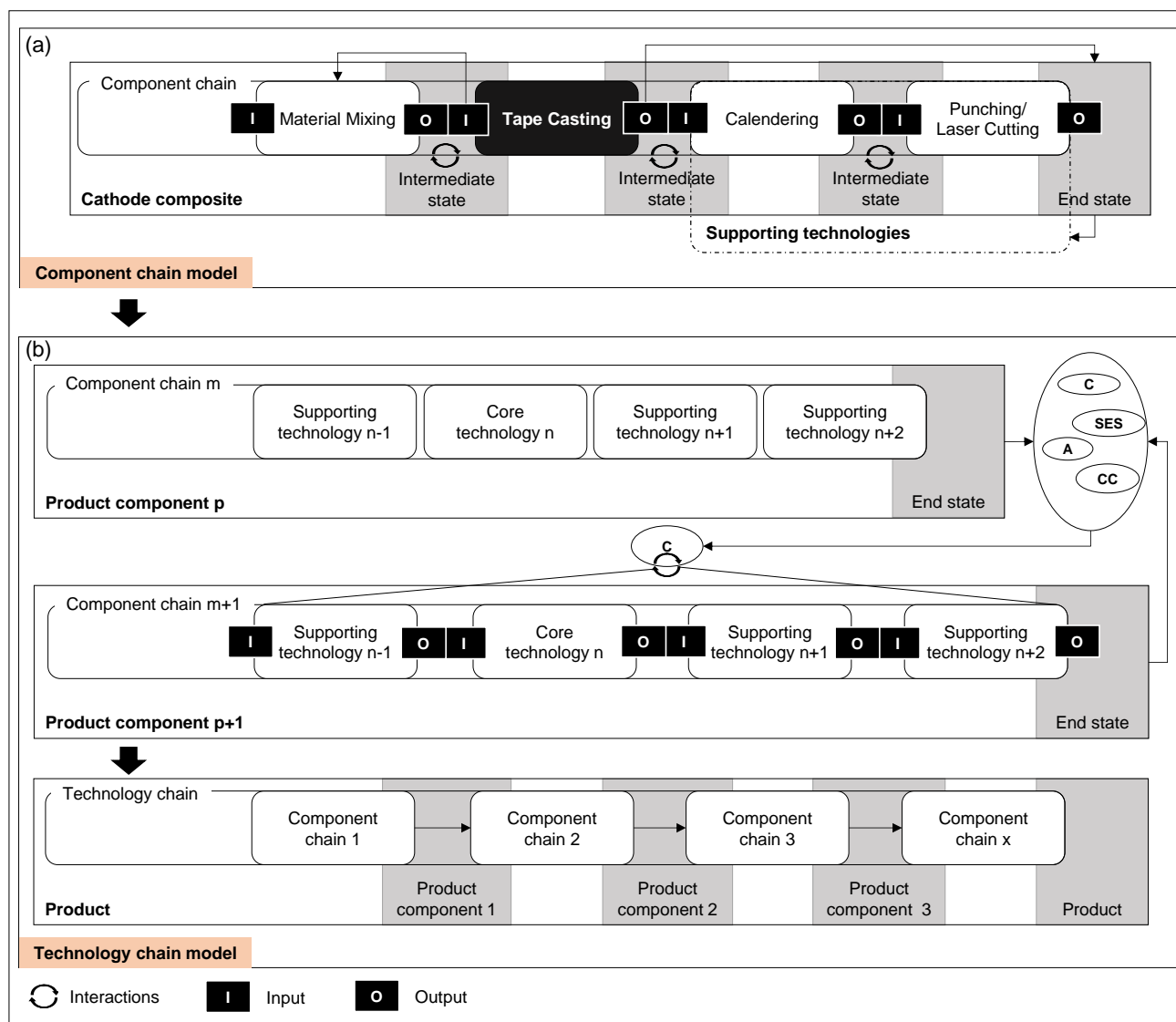


Figure 6. Schematic procedure of a) component chain generation for sulfidic cathode composite and b) generic technology chain generation. Components of the cell: CC = current collector, SES = solid electrolyte separator, A = anode, C = cathode composite.

requirements are fulfilled, the technology chain is generated using the technology model. A subsequent analysis of the generated technology chain is necessary and structural measures can be used to adjust the technology chain before final selection.

First, the core technologies have to be assigned to suitable substrates. Substrates are needed to build up thin layers, which are not mechanically stable by itself. Cell layers such as the cathode composite can be coated directly with the SES and is used as a substrate. Another possibility is to coat the SES on a substrate, which is then removed. For the characterization of the substrates, criteria such as the thermal stability, surface roughness, and porosity of the layers are defined. In the next step, the criteria of the substrates are reviewed and compared with the properties of the technologies used in the component chain. The exclusion of component chains for sulfidic ASSBs was completed in the following example. The technologies PLD and ADM were excluded for the sulfidic cell due to their high process temperatures, which are not compatible when using a polymer substrate. In contrast to the concept of a bipolar stacking,^[90] these technologies are not affected in a parallel stacking because the aluminum and copper collector foils, used as substrates, can resist the temperatures. After the generation of the technology chain, adjustments can be necessary. In the present case, the joint cutting of half cells with the lithium anode bears the risk of metallic cross-contamination. As a measure, the dimensions of the anode layer can be reduced so that only the collector foils are cut. Otherwise, the anode layer on top of the collector foil must be cut to size separately.

5.1. Technology Chain Model

The technology chain model associates specific technologies to each technology function. It has input and output ports as the interfaces to the system boundary, for example, materials and components entering the process (further inputs could be, for instance, solvents, pore formers, carrier gases, carrier tapes) and the ASSB as final output. In the exemplary technology model shown in Figure S5, Supporting Information, only the superordinate technology functions (sheet fabrication, stack assembly, cell assembly) are shown, represented by the corresponding component chains. Each component chain in turn has input and output ports which can be connected to other ports. The technology chain model consists of different component chains, which are then combined to build final technology chains (Figure S9, Supporting Information).

Generated technology chains for this specific cell design are shown in Figure 7. Beginning with the mixing step to fabricate starting powders or slurries, techniques such as planetary mixing or ball milling are identified. For the layer fabrication, suitable technologies are wet coating processes or powder processes, e.g., ADM or PLD. As mentioned earlier, the latter can only be applied, if a parallel stack (p) is produced. In this case, a cathode-supported cell was selected, where the SES is coated directly onto the cathode composite. If a protective layer is needed, it can be either applied onto the SES (Figure 7) or the anode using evaporation or tape casting. In addition, first considerations about the assembly of ASSB cells are shown. Conventional die cutting of cell layers seems to be not applicable

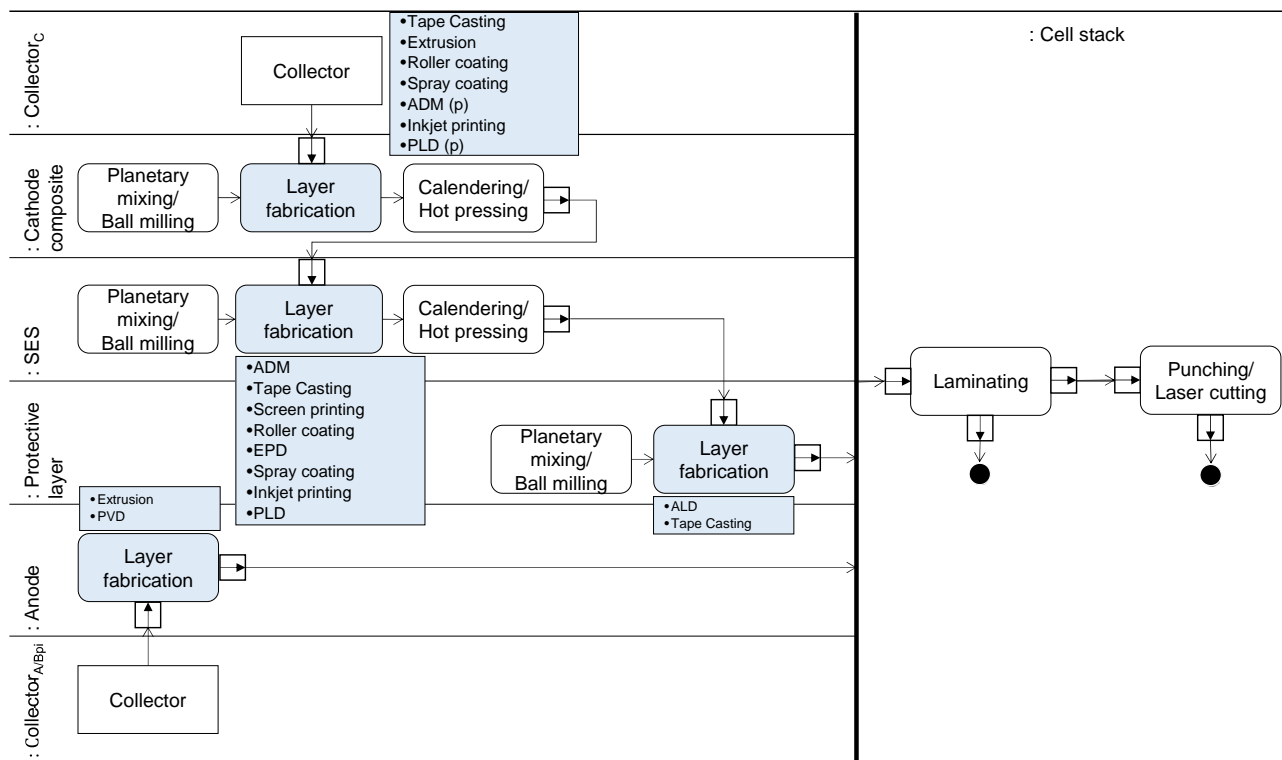


Figure 7. Internal block diagram for technology chains for sulfidic cells with a cathode-supported cell design. The focus in this figure is set on alternatives for layer fabrication processes (colored fields).

for all novel materials. Mechanical separation of sulfides might be sufficient, whereas oxides or lithium metal seem to need a laser-cutting process to be separated adequately. After coating the anode (lithium metal) on the current collector, all cell layers can be laminated and then cut to size. The sequence of cutting and assembling a cell stack varies depending on the cell design (bipolar collector or parallel stacking) and solid electrolyte material.

5.2. Technology Profiles

As a result of previous models, a technology profile was created for every process (Figure 8). It contains a sketch, a description of the functional principle, the generated technology chain for a half cell (cathode composite with SES) as well as some alternative technologies.

6. Evaluation of Technology Chains

At first, a rough evaluation of all technologies in the respective technology chain takes place (Figure S10, Supporting Information). A defined number (m) of criteria (C) is chosen to evaluate the feasibility of the technology chains. To consider the company's strategic direction, a weighting of those criteria is used. If the user has no preferences, all values are weighted equally and then normalized to the number of technologies in the chain (n) and calculated by multiplication with the weightings (W). The sum of the individual values in Equation (2) gives the technology chain value (T_C). This value is decisive for the choice

of technology chains. Afterward, a design review can be carried out to adjust the product to possible limitations of technologies. The design review gives the user a reinsurance, whether the cell design is possible to manufacture. Here, parameters as, for instance, the layer thickness can be revised to consider the mechanical strength of the layer during the fabrication. Also other parameters, which differ from the properties of conventional LIB, such as the reduction of the porosity to a minimum or interfacial adhesion between solid layers might arise challenges in the production. The design review is needed to ensure the producibility with the selected technologies.

$$\sum_1^m \frac{\sum_1^n C_n \times W_{C_n}}{n} = T_C \quad (2)$$

To evaluate technology chains, further analysis is required. Within the frame of this publication, the generated technology chains were automatically evaluated and selected by using a specifically developed tool with a user interface. The technology chains are exemplarily prioritized by the criteria of technology maturity (TM), quality (Q), and throughput (T). Based on Heimes,^[91] technology chains are sorted by weighting different criteria: A technology-oriented chain focuses on the technology maturity value; a product-oriented technology chain prioritizes the quality of the chain measured in terms of the product quality; a process-oriented process chain concentrates on the throughput.

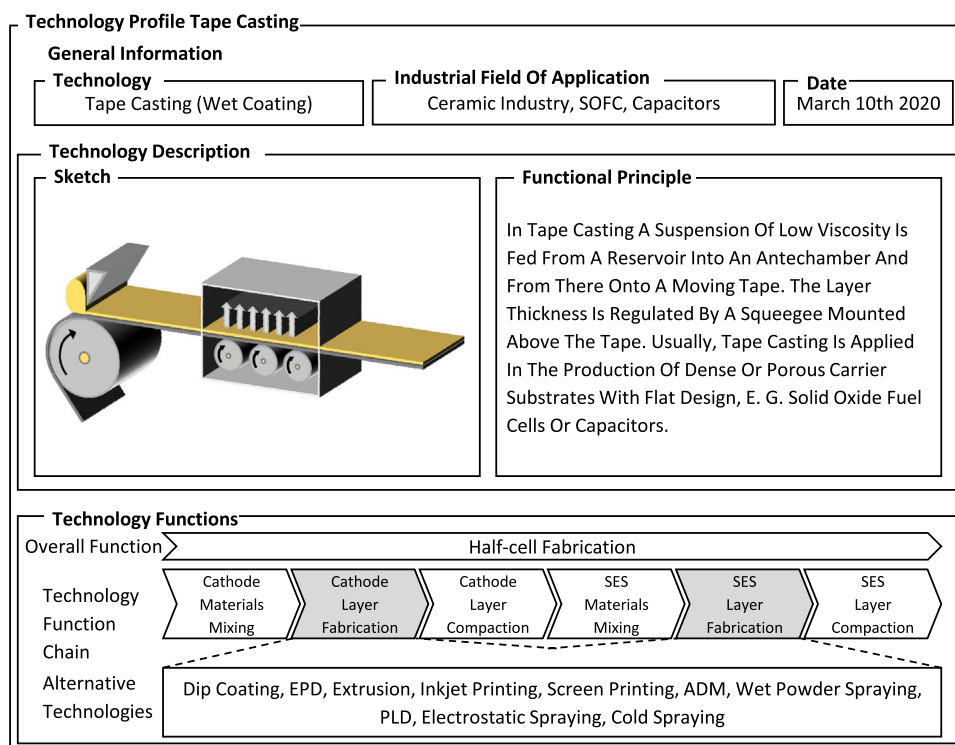


Figure 8. Exemplary technology profile for tape casting with overview on functional principle, allocation to technology function, and alternative technologies.^[88,95,96]

7. Application of the System Model Using a Technology Generation Tool

The user interface consists of defining product and process features, which are chosen for a sulfidic cell as following: After selecting a cell housing, the user can determine the cell format, the SES, and cathode material. Finally, a parallel or bipolar interconnection of the cell needs to be chosen (Figure S12, Supporting Information). If necessary, the SES content in the cathode composite (default value: 30 vol%)^[92] and anode (default value: 0 vol% for lithium metal) as well as the ratio of cathode capacity to anode capacity (default value: 1.1) can be changed manually. The layer thickness of the electrolyte is set to 50 μm by default.^[93] With these entries, the user has the possibility to set a self-selected cell design. The information on the company's production environment is important for prioritizing the technology chains. The production volume can be roughly divided into small (≈ 20 MWh), medium (≈ 200 MWh), and large (≈ 2 GWh) series individually.^[91] The resulting output from the tool is a selection of technology chains, which illustrate production paths for a defined cell design under consideration of corporate strategic preferences. The specifications for the sulfidic cell are listed in Table 1. All other values are set by default (see earlier).

After generating all possible chains (Figure 7), the automated tool uses the input with cell data to browse every registered technology with its values for quality, time, and maturity. The weighting of these values allows the tool to sort and rank technologies for each process step. As a result, technology chains with the highest T_C -value are listed. Exemplary, the four best evaluated chains to fabricate a sulfidic solid electrolyte are using tape casting and screen printing as core technologies and differ mainly in the layer cutting process step (Figure 9). Although punching of ASSB layers shows advantages in maturity and time, laser cutting is preferably used to gain a higher quality. In total, technology chain 1 (TC1) with tape casting and punching has a maturity

Table 1. Selection of product and production features for the ASSB. Plug-in hybrid electric vehicle (PHEV2) cell format has dimensions of $91 \times 148 \times 26.5$ mm for prismatic hard cases^[97]; LSPS is a common sulfidic solid electrolyte material as a composition of $\text{Li}_2\text{S-P}_2\text{S}_5$ ^[38]; HE-NMC as lithium-rich cathode active material $x\text{Li}_2\text{MnO}_3 (1-x)\text{LiMO}_2$ ($M = \text{Mn, Co, Ni}$)^[98]; LiPON as oxide electrolyte, mostly used in thin-film batteries or as protective layer.^[99]

Features	
Cell housing	Prismatic hard case
Cell format	PHEV2
SSE material	LSPS
Cathode active material	HE-NMC
Catholyte (SE in cathode)	LSPS
Protective layer	LiPON ^[21]
Cathode layer thickness	150 μm
Stacking	Bipolar ^[90]
Production volume	2 GWh a^{-1}
Technology chain type	Process-oriented

distribution around 90% (Figure 9a), whereas TC3 with screen printing and punching is around 85% (Figure 9c). Both technologies and, therefore, the chains suffer from insufficient experience for ASSBs. This results from the fact that there is no industrial fabrication of ASSBs using those technologies at the moment. Nevertheless, wet coating technologies seem to be most promising for forming thin layers in ASSBs. The application of punching or laser cutting for sulfidic cells will be decided according to the demands on the cut edges, as laser cutting promises a higher quality (Figure 9b,d).

8. Discussion

The relationship between product and production technology to upscale ASSB production is very complex. The model shows an approach how to identify and detail the complexity of these interrelations. The shown data in the article are a part of the work that was done to create the system model or were calculated and presented in previous work^[19–21,94] of the research group. All data discussed is shown as an example on how to describe the complex interactions between the product and the processes. A range of data can be added to the model, i.e., the model can be extended at any time, as more properties of the solid electrolyte will be investigated in the next months and years. The development of the ASSB research would be underlined by a growing model, which then will include more detailed information in the future. To date, all applicable data and correlations have been tried to be considered. The novelty of the results lies in the holistic view of the system model. The modeling allows finding the right production processes adapted to the respective company independent of the used materials and cell designs. The model functions as an automated tool that can be used to assemble different technology chains based on a user selection. The structure of the model itself works for the shown cell designs and chosen processes very well. The differentiation according to product quality, throughput, or technological maturity in the technology chains also makes it possible to intercept application-specific production requirements. Each SSB can be used for a wide range of applications and in these fields, the production facilities and standards differ massively.

9. Conclusion

As a result, a method for modeling product and process interdependencies in the ASSB production was developed. In addition to structural models, also the behavior of the production processes was analyzed. Conclusions can be drawn to elaborate technology chains for ASSBs ranked by individual company criteria. The strong linkage of the variables related to the cell design and the technologies shows that a simultaneous development of product and process for the ASSB production is necessary. Accordingly, major changes will be necessary for the production of an ASSB compared with conventional LIB production. Exemplarily, the method was carried out for a sulfidic ASSB. Choosing a process-orientation, by means of a high throughput, a process route consisting of planetary mixing, tape casting, calendaring, laminating, and punching layers would fit the companies' strategy most. The results shown in this article shall

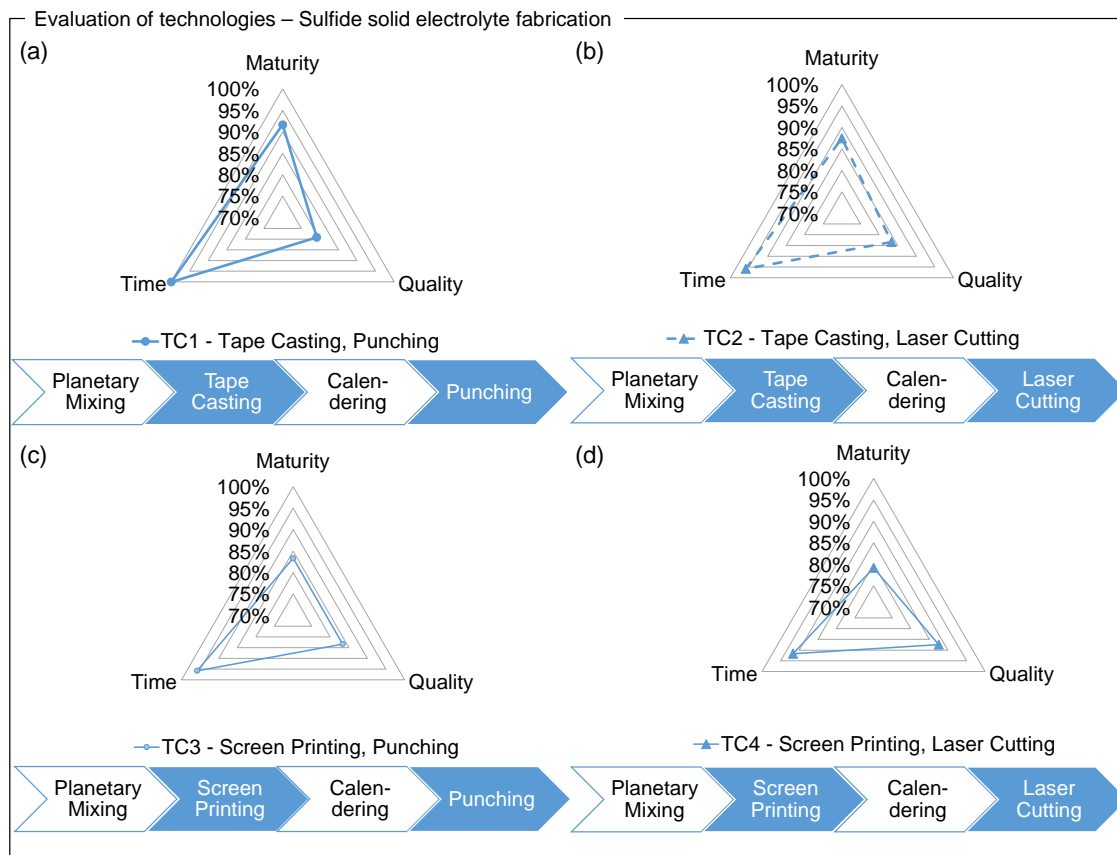


Figure 9. Evaluation of maturity, time, and quality between technology chains for the sulfidic solid electrolyte fabrication. a) Technology chain (TC1) with tape casting and punching. b) Technology chain (TC2) with tape casting and laser cutting. c) Technology chain (TC3) with screen printing and punching. d) Technology chain (TC4) with screen printing and laser cutting. The value 100% refers to a standardized process which is already used in mass production. Given values for the maturity are averages from a distribution in the Monte Carlo Simulation.

support cell manufacturing companies to take a next step toward the development of safer batteries and close the gap from laboratory to an ASSB industrial production.

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.

Keywords

all-solid-state batteries, all-solid-state battery production, production planning, sulfide solid electrolytes, system modeling

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