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Institut für Werkzeugmaschinen und Betriebswissenschaften (*iwb*)

**Strategic Technology Planning  
for the Production of All-Solid-State Batteries**

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## Editors' Preface

Production engineering is crucial for the advancement of our industrial society because the performance of manufacturing companies depends heavily on the equipment and resources employed, the production processes applied, and the established manufacturing organization. A company's full potential for corporate success can only be reached by optimizing the interaction between humans, operational structures, and technologies. Being able to remain competitive while balancing the varying and often conflicting priorities of complexity, cost, time, and quality requires constant thought, adaptation, and the development of new manufacturing structures. Thus, there is an essential need to reduce the complexity of products, manufacturing processes, and systems. Yet at the same time it is also vital to gain a better understanding and command of these aspects.

The objective of the research activities at the Institute for Machine Tools and Industrial Management (*iwb*) is to continuously improve product development and manufacturing planning systems, manufacturing processes, and production facilities. A company's organizational, manufacturing, and work structures, as well as the underlying systems for order processing, are developed under strict consideration of employee-related requirements. Although an increasing degree of automation is unavoidable, labor will remain an important component in production processes. Thus, questions concerning the optimization of human involvement in the Idea-to-Offer process are of utmost importance.

The volumes published in this book series collate and report the results from the research conducted at the *iwb*. Research areas covered stretch from the design and development of manufacturing systems to the application of technologies in manufacturing and assembly. The management and operation of manufacturing systems, quality assurance, availability, and autonomy are overarching topics, which affect all areas of our research. In this series, the latest results and insights from our application-oriented research are published. These will foster an improvement in the transfer of knowledge between universities and the wider industrial sector.

*Gunther Reinhart*

*Michael Zäh*



## **Abstract**

A successful transition towards renewable energies and electric vehicles requires efficient and cost-effective energy storage systems. Compared to conventional lithium-ion batteries (LIBs), the all-solid-state battery (ASSB) offers the potential for higher energy densities with improved safety. Although promising results have been achieved at the laboratory level, there is still little empirical know-how on upscaling. The objective of the present work is, therefore, to create a broad knowledge base for the industrial production of ASSBs by systematically analyzing challenges and solutions for the successful fabrication of ASSBs.

To this end, the current state of research on ASSBs will first be summarized with a special focus on the corresponding manufacturing processes. Subsequently, approaches for the generation and evaluation of technology chains within the framework of strategic technology planning will be critically examined with regard to their suitability for the given research problem. Based on the identified need for action, a methodology will be presented specifically for the underlying research questions.

A hierarchical ASSB product model enables the systematic analysis of production-relevant characteristics and the identification and evaluation of suitable production technologies and technology chains. Based on the results of an expert survey, challenges and requirements for the industrial production of ASSBs are identified and corresponding processing routes are derived. A systematic assessment of alternative manufacturing technologies in the production of LIBs, fuel cells, and ceramic capacitors enables a top-down calculation of manufacturing costs by means of learning curve effects. In order to enable a direct comparison with conventional LIB production, a bottom-up model is presented to calculate the cost development along the process chain for different scenarios. This allows to identify critical process steps in the production of ASSBs.

Finally, the results are verified by applying the methodology together with an international battery manufacturer. In this way, the economic and technical benefits of the methodology are demonstrated. The concluding critical reflection enables an outlook on improvement potentials and further research activities. In

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summary, the results of this work make a valuable contribution to the scale-up of ASSBs and provide a profound decision-making aid for both applied research and industry.

## Kurzfassung

Eine erfolgreiche Energie- und Mobilitätswende hin zu erneuerbaren Energien und Elektrofahrzeugen erfordert effiziente und kostengünstige Energiespeicher. Die Festkörperbatterie bietet im Vergleich zu konventionellen Lithium-Ionen-Batterien das Potenzial für höhere Energiedichten bei verbesserter Sicherheit. Obwohl auf Laborebene bereits vielversprechende Ergebnisse erzielt werden konnten, gibt es bisher nur wenig Erfahrungswissen zur Hochskalierung. Die Zielsetzung der vorliegenden Arbeit ist somit die Schaffung einer breiten Wissensbasis zur industriellen Herstellung von Festkörperbatterien, indem Herausforderungen und Lösungsansätze für eine erfolgreiche Produktion von Festkörperbatterien systematisch analysiert werden.

Hierzu wird zunächst der aktuelle Wissensstand zu Festkörperbatterien mit einem besonderen Augenmerk auf entsprechende Herstellungsverfahren zusammengefasst. Darauf aufbauend werden Ansätze zur Generierung und Bewertung von Technologieketten im Rahmen der strategischen Technologieplanung hinsichtlich ihrer Eignung für die gegebene Fragestellung kritisch beleuchtet. Basierend auf dem aufgezeigten Handlungsbedarf wird eine Methodik speziell für die vorliegende Problemstellung präsentiert.

Ein hierarchisches Produktmodell für Festkörperbatterien ermöglicht die systematische Erfassung von herstellungsrelevanten Merkmalen und eine Identifikation und Bewertung geeigneter Produktionstechnologien und Technologieketten. Basierend auf den Ergebnissen einer Expertenbefragung werden Herausforderungen und Anforderungen an eine industrielle Herstellung von Festkörperbatterien ermittelt und entsprechende Verfahrensrouten abgeleitet. Eine systematische Bewertung von alternativen Fertigungstechnologien in der Herstellung von Lithium-Ionen-Zellen, Brennstoffzellen und Keramikkondensatoren ermöglicht eine Top-down-Berechnung der zu erwartenden Herstellkosten mittels Lernkurven-Effekten. Um einen direkten Vergleich mit der konventionellen Batterieproduktion zu ermöglichen, wird anschließend ein Bottom-up-Modell präsentiert, um die Kostenentstehung entlang der Prozesskette für unterschiedliche Szenarien zu berechnen und somit kritische Prozessschritte in der Herstellung von Festkörperbatterien zu identifizieren.

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Schließlich erfolgt eine Verifizierung der entwickelten Methodik anhand eines Anwendungsbeispiels gemeinsam mit einem internationalen Batteriehersteller. Dadurch kann der Nachweis für einen wirtschaftlichen und technischen Mehrwert erbracht werden. Die kritische Reflexion der Ergebnisse ermöglicht abschließend einen Ausblick auf Verbesserungspotenziale und weitere Forschungsaktivitäten. Die Ergebnisse dieser Arbeit leisten somit einen wertvollen Beitrag zur Hochskalierung von Festkörperbatterien und ermöglichen eine fundierte Entscheidungshilfe sowohl für die angewandte Forschung als auch für die Industrie.

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## Abbreviations

<b>ADM</b>	aerosol deposition method
<b>AHP</b>	Analytic Hierarchy Process
<b>ALD</b>	atomic layer deposition
<b>ASSB</b>	all-solid-state battery
<b>BatPaC</b>	Battery Performance and Cost model
<b>BEV</b>	battery electric vehicle
<b>CO<sub>2</sub></b>	carbon dioxide
<b>CVD</b>	chemical vapor deposition
<b>DFMA</b>	Design for Manufacture and Assembly
<b>DMM</b>	Domain Mapping Matrix
<b>DRM</b>	Design Research Methodology
<b>DSM</b>	Design Structure Matrix
<b>EPD</b>	electrophoretic deposition
<b>FELIZIA</b>	<i>Festelektrolyte als Enabler für Lithium-Zellen in automobilen Anwendungen</i> (Engl. <i>Solid electrolytes as enabler for lithium cells in automotive applications</i> ), funded by the Federal Ministry of Education and Research (Bundesministerium für Bildung und Forschung), grant number 03XP0026I
<b>HE-NMC</b>	$x \text{Li}_2\text{MnO}_3 \cdot (1 - x) \text{LiMO}_2$
<b>HVOF</b>	high velocity oxygen fuel spraying
<i>iwb</i>	Institute for Machine Tools and Industrial Management
<b>LAGP</b>	$\text{Li}_{1+x}\text{Al}_x\text{Ge}_{2-x}(\text{PO}_4)_3$
<b>LATP</b>	$\text{Li}_{1+x}\text{Al}_x\text{Ti}_{2-x}(\text{PO}_4)_3$
<b>LBO</b>	$\text{Li}_3\text{BO}_3$
<b>LCO</b>	$\text{LiCoO}_2$
<b>LFP</b>	$\text{LiFePO}_4$
<b>LGPS</b>	$\text{Li}_2\text{S}-\text{GeS}_2-\text{P}_2\text{S}_5$
<b>LIB</b>	lithium-ion battery
<b>LIC</b>	lithium-ion cell
<b>Li-ion</b>	lithium-ion
<b>LISICON</b>	Lithium SuperIonic CONductors
<b>LIPON</b>	$\text{Li}_x\text{PO}_y\text{N}_z$

## Abbreviations

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<b>LLTO</b>	$\text{Li}_{0.5}\text{La}_{0.5}\text{TiO}_3$
<b>LLZ</b>	$\text{Li}_x\text{La}_y\text{Zr}_2\text{O}_{12}$
<b>LLZO</b>	$\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$
<b>LLZ:Ta</b>	$\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$
<b>LLZ:Te</b>	$\text{Li}_{6.5}\text{La}_3\text{Zr}_{1.75}\text{Te}_{0.25}\text{O}_{12}$
<b>LMB</b>	lithium metal battery
<b>LMO</b>	$\text{LiMn}_2\text{O}_4$
<b>LNMO</b>	$\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$
<b>LSiPSCI</b>	$\text{Li}_{9.54}\text{Si}_{1.74}\text{P}_{1.44}\text{S}_{11.7}\text{Cl}_{0.3}$
<b>LPS</b>	$\text{Li}_2\text{S}-\text{P}_2\text{S}_5$
<b>LPSCI</b>	$\text{Li}_6\text{PS}_5\text{Cl}$
<b>LPSI</b>	$\text{Li}_2\text{S}-\text{P}_2\text{S}_5-\text{LiI}$
<b>LPSX</b>	$\text{Li}_6\text{PS}_5\text{X}$
<b>LTO</b>	$\text{Li}_4\text{Ti}_5\text{O}_{12}$
<b>MCS</b>	Monte Carlo Simulation
<b>MLCC</b>	multi layer ceramic capacitor
<b>NASA</b>	National Aeronautics and Space Administration
<b>NCA</b>	$\text{Li}(\text{Ni}_{1-x-y}\text{Co}_x\text{Al}_y)\text{O}_2$
<b>NMC</b>	$\text{Li}(\text{Ni}_{1-x-y}\text{Mn}_x\text{Co}_y)\text{O}_2$
<b>NMC811</b>	$\text{Li}(\text{Ni}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1})\text{O}_2$
<b>NASICON</b>	NAtrium SuperIonic CONductors
<b>OEM</b>	original equipment manufacturer
<b>PEO</b>	poly(ethylene oxide)
<b>PLD</b>	pulsed laser deposition
<b>ProFeLi</b>	<i>Produktionstechnik für Festkörperzellen mit Lithium-Metall-Anode (Engl. Production technology for all-solid-state batteries with lithium metal anode), funded by the Federal Ministry of Education and Research (Bundesministerium für Bildung und Forschung), grant number 03XP0184I</i>
<b>PV</b>	photovoltaics
<b>PVD</b>	physical vapor deposition
<b>QFD</b>	Quality Function Deployment
<b>SD</b>	staff day
<b>SE</b>	solid electrolyte
<b>SES</b>	solid electrolyte separator

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<b>SOFC</b>	solid oxide fuel cell
<b>SPS</b>	spark plasma sintering
<b>SSB</b>	solid-state battery
<b>SWOT</b>	Strenghts, Weaknesses, Opportunities, Threats
<b>TRL</b>	Technology Readiness Level
<b>TUM</b>	Technical University of Munich



## List of Symbols

$A$	area (in $\text{cm}^2$ )
$C$	(discharge) capacity (in Ah)
$c_{\text{spec},c}$	cathode active material specific capacity (in mAh/g)
$c_{\text{spec},a}$	anode active material specific capacity (in mAh/g)
$C_{\text{area}}$	area specific capacity (in mAh/cm <sup>2</sup> )
$d_{\text{el}}$	electrode thickness (in $\mu\text{m}$ )
$d_{\text{s}}$	separator thickness (in $\mu\text{m}$ )
$E$	energy (in Wh)
$E_{\text{spec}}$	specific energy (in Wh/kg)
$E_{\text{vol}}$	energy density (in Wh/L)
$I$	current (in mA)
$J$	specific current (in mA/cm <sup>2</sup> )
$M_{\text{c}}$	cathode active material areal loading (in g/cm <sup>2</sup> )
$M_{\text{a},c}$	anode/cathode active material areal loading (in g/cm <sup>2</sup> )
$M_{\text{i}}$	areal weight of individual cell components (in g/cm <sup>2</sup> )
$M_{\text{p}}$	areal weight of passive electrode components (in g/cm <sup>2</sup> )
$R$	Ohmic resistance (in $\Omega$ )
$\bar{V}$	average discharge voltage (in V)
$\Delta V$	voltage drop (in V)
$x_{\text{p}}$	electrode porosity (in vol%)
$\sigma_{\text{i}}$	ionic conductivity (in S/cm)
$\rho_{\text{a},c}$	anode/cathode active material density (in g/cm <sup>3</sup> )
$\rho_{\text{i}}$	density of individual cell components (in g/cm <sup>3</sup> )
$\rho_{\text{p}}$	density of passive electrode components (in g/cm <sup>3</sup> )



# 1 Introduction

*"We became human beings by working together and helping one another [...]. What it takes to reverse global warming is one person after another remembering who we truly are."* PAUL HAWKEN, Drawdown

## 1.1 Motivation

The transfer from fossil fuels to energy from renewable sources is an inevitable step to reduce the ecological and social hazards caused by global warming and pollutant emissions. Efficient means of energy storage are required to buffer fluctuations caused by wind and solar power generation and to enable emission free mobility. Especially with the transfer from combustion engine to electrically driven vehicles, a shift in the value-added chain towards the energy storage system and a sharp rise in demand for large-format lithium-ion cells (LICs)<sup>1</sup> is to be expected in the upcoming decade (THIELMANN 2017). This causes challenges, but also chances for automotive companies and suppliers: Established LIC manufacturers and also newcomers on the market are planning to implement new LIC production facilities with capacities of several tens of GWh/year. Therefore, a strong competition and price decline are to be expected on the global market for large-format LICs (NYKVIST & NILSSON 2015; SCHMIDT ET AL. 2017).

In a conventional LIC, the liquid electrolyte soaks the porous electrodes (anode and cathode) and separator to enable ion transfer during charge and discharge (LEUTHNER 2018, p. 14) (figure 1.1, left). By replacing the liquid electrolyte with an ion conducting **solid electrolyte (SE)**, many of the issues currently associated with LICs could potentially be overcome: An **all-solid-state battery (ASSB)** is a battery<sup>2</sup> which does not contain any liquid components<sup>3</sup> and

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<sup>1</sup> A *lithium-ion cell* is comprised of one or multiple galvanic elements, typically encapsulated into a cylindrical or prismatic hardcase or pouch packaging (WOEHRLE 2018, p. 105). In this context, *large-format* refers to dimensions according to DIN 91252.

<sup>2</sup> The term *battery* is used ambiguously in literature for galvanic elements up to battery systems/packs and will be used as a superordinate term in this context. The term *all-solid-state battery* thus also accommodates the possibility for innovative cell concepts enabled by the use of SEs (cf. section 2.1).

<sup>3</sup> This is in contrast to so called quasi-solid-state batteries (KELLER ET AL. 2018), where liquids may be added to enhance ionic conductivity and reduce interface resistances, e.g. by the use of gel polymer electrolytes (cf. section 2.2).

# 1 Introduction

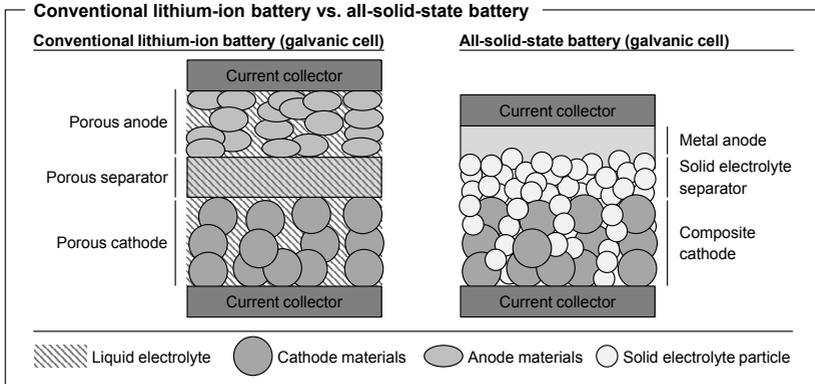


Figure 1.1: Schematic illustration of a conventional lithium-ion battery (galvanic cell, left) and an all-solid-state battery with a metal anode (galvanic cell, right).

uses a solid electrolyte separator (SES) to enable ion transfer and ensure electric isolation between the electrodes (figure 1.1, right) (HOOPER & TOFIELD 1984). The elimination of flammable liquid components is expected to enhance battery safety (INOUE & MUKAI 2017). Furthermore, a superior energy density could be achieved by facilitating the use of different electrode materials (e.g. a lithium metal anode, cf. section 2.2), and cell designs (e.g. bipolar stacking, cf. section 2.1). Additionally, the omission of the electrolyte filling process could potentially simplify manufacturing processes (GRAPE 2015, p. 17). Therefore, ASSBs are promising candidates for a disruptive change which could facilitate the market entry for cell manufacturers. The associated material innovations and the need for adapted production technologies<sup>4</sup> (cf. next section) could also serve as a chance for material and component suppliers, machine and systems engineering, and original equipment manufacturers (OEMs).

<sup>4</sup> The term *technology* describes “the specific know-how, skills, methods and facilities necessary for the deployment of engineering and scientific knowledge” (TSCHIRKY 1994). Depending on the application, a differentiation is typically made between product and production technologies (ibid.). The focus of this thesis is on primary (i.e. value-adding) production technologies: *Process technologies* deal with the synthesis and transformation of substances with undefined shape (raw materials). *Manufacturing technologies* serve for fabrication and processing of bodies with a geometrically defined shape and are categorized as shaping, forming, separating, joining, coating, and properties changing according to DIN 8580. *Assembly technologies* according to VDI 2803 comprise joining technologies for the assemblage of bodies with a geometrically defined shape, as well as secondary tasks, such as handling and quality checks which are not within the scope of this thesis.

## 1.2 Problem Statement

Despite the expected advantages of ASSBs, several issues have come up with the use of a SE: While in conventional LICs the liquid electrolyte wets the porous electrodes and separators to form a large surface for ion exchange (KNOCHÉ ET AL. 2016), the ion transfer in an ASSB is limited to the solid-solid interfaces of the SE particles and the electrode active materials<sup>5</sup> (“point contacts”) (HU 2016). Therefore, intimate contact and low porosities are required to ensure low interface resistances (LUNTZ ET AL. 2015), which is a prerequisite for fast charging and high power (KATO ET AL. 2016c). Additionally, concerns arise due to limited electrochemical stability of some SEs (ZHU ET AL. 2015), necessitating the modification of interfaces or insertion of additional protective layers (OHTA ET AL. 2006; HAN ET AL. 2016). Moreover, the porous media in conventional LICs can to some extent compensate for volume changes of the electrode active materials during battery operation, while the low porosities and unfavorable elastic properties in ASSBs lead to limited mechanical stability and non-successive degradation (KOERVER ET AL. 2017; LIU ET AL. 2017).

These issues also imply possible manufacturing challenges and hinder a simple transfer of production processes from conventional LICs, as illustrated in figure 1.2: The technology chain<sup>6</sup> for conventional LIC production comprises the manufacturing of the electrodes, the cell assembly, and the cell formation and final test. At first, the electrode materials (active materials, conducting agents, binders) are mixed and dispersed with a solvent before wet coating onto a current collector foil and evaporation of the solvent (BRODD & TAGAWA 2002, 272ff. GÜNTHER ET AL. 2016). After calendaring to adjust the layer properties (GÜNTHER ET AL. 2019), the resulting electrode rolls are typically slitted to smaller coils (PETTINGER ET AL. 2018, p. 215). During cell assembly, the electrodes are cut to fit the cell format (KURFER ET AL. 2012) and joined with the separator in a stacking or winding process (REINHART ET AL. 2013; PETTINGER ET AL. 2018, 215ff.). This is followed by welding of the current collector tabs,

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<sup>5</sup> An *active material* in this context is defined as an anode or cathode material whose main purpose it is to store and release lithium-ions (Li-ions), cf. section 2.2 (LEUTHNER 2018, p. 14).

<sup>6</sup> *Technology chains* can be defined as “abstract, manufacturing facility independent combinations of production technologies in a defined sequence. They allow for the entire technological value-added chain to be taken into account and interdependencies between individual technologies to be considered” (KLOCKE ET AL. 2000).

# 1 Introduction

packaging into the cell housing, and electrolyte filling (KNOCH & REINHART 2015; PETTINGER ET AL. 2018, 217f.). In the final steps, the cells are subject to elaborate charge and discharge processes (formation), degassed, aged, and classified in an end-of-line quality check (BRODD & TAGAWA 2002, p. 279).

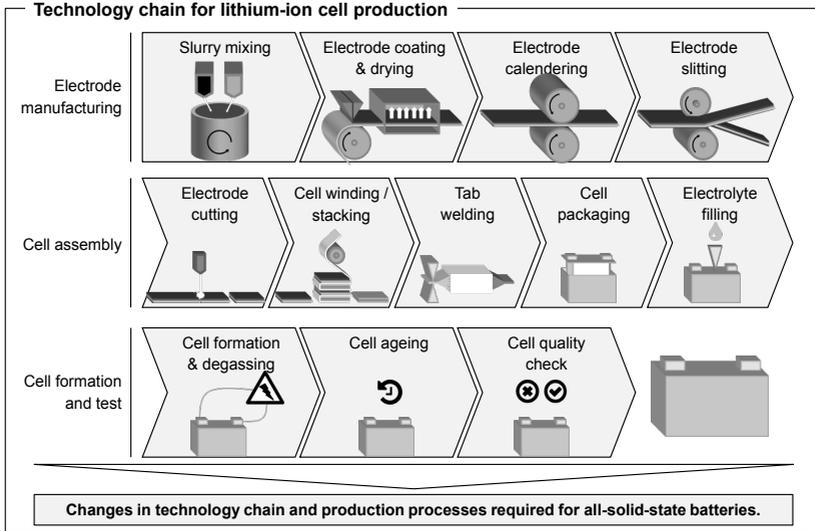


Figure 1.2: Illustration of a typical technology chain for LIC production from materials mixing to the final battery cell. Due to the different properties of ASSBs, changes will be required in the production processes.

The replacement of the separator and liquid electrolyte by a SE, the required low porosities, and the aforementioned issues concerning the interfaces in ASSBs necessitate a rethinking of these processes: In order to allow for sufficient ionic percolation inside the electrodes, it may be necessary to prepare composite electrodes with a combination of SE particles and electrode materials (HOOPER & TOFIELD 1984). Manufacturing of the SES and, potentially, also the protective layer(s) requires an additional coating or shaping process (INADA ET AL. 2009; GRAPE 2015, p. 16). High pressure or temperature may be required to achieve sufficiently low porosities and low interface resistances in composite electrodes and SES (SAKUDA ET AL. 2013; YAMAMOTO ET AL. 2017). The use of new materials and cell designs may also call for adaption of cutting, stacking, welding, and packaging processes (SCHNELL ET AL. 2018). As described above,

no electrolyte filling will be required for ASSBs, and—depending on the materials combination—the formation procedure needs to be adjusted (GRAPE 2015, p. 17). Due to the limited chemical stability of some SE materials, processes may need to be carried out under inert gas (MURAMATSU ET AL. 2011).

The above described challenges lead to high uncertainty regarding product and production planning (KERMAN ET AL. 2017), resulting in a high risk and potentially high initial cost for ASSB production: From a product perspective, the so far unresolved issues currently hinder a detailed specification of the ASSB design, i.e. materials selection and cell dimensions. The issues associated with ASSB fabrication could overstrain the capabilities of established production technologies and, therefore, call for innovations and development of technologies with a low degree of maturity<sup>7</sup>. However, a competitive advantage in ASSB production can only be achieved by numbers of scale, especially with regard to the strong price decline of conventional LIBs during the recent years (NYKVIST & NILSSON 2015; SCHMIDT ET AL. 2017). Projections for the actual demand and performance development of large-format LICs vary strongly (THIELMANN 2017), hindering precise planning of the quantities of ASSBs to be produced. Despite the great opportunity to successfully enter the turbulent LIB market with a disruptive (product) technology such as the ASSB, industrial companies must carefully consider how to spend their limited resources in a long-term planning horizon (SCHULTE-GEHRMANN ET AL. 2011, p. 58). This necessitates profound decision making within the activities of strategic technology management<sup>8</sup>, in particular concerning strategic technology planning<sup>9</sup>, as illustrated in figure 1.3 (REINHART ET AL. 2011; SCHUH ET AL. 2011c, 171ff.).

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<sup>7</sup> The *technology maturity* represents the degree of development of a technology (REINHART & SCHINDLER 2010). Usually, the higher the degree of maturity, the lower the risk associated with a technology's utilization (BROUSSEAU ET AL. 2009).

<sup>8</sup> *Technology management* "links engineering, science, and management disciplines to plan, develop, and implement technological capabilities to shape and accomplish the strategic and operational objectives of an organization" (HERINK ET AL. 1987, p. 9). *Strategic technology management* is "planning, organising, leading and controlling of technological activities [...] to contribute to formulation and execution of company's basic, long-term goals and objectives [...]" (SAHLMAN & HAAPASALO 2009).

<sup>9</sup> *Strategic technology planning* is intended to maintain and expand the competitiveness of a company (METZ 1996). The purpose is to synchronize knowledge regarding products, technologies, and their relations at an early stage in order to identify and evaluate technology alternatives during product development (MILBERG & MÜLLER 2007; MÜLLER 2008, p. 98). This is in contrast to operational technology planning which rather focuses on short-term planning activities such as the allocation of production steps to manufacturing resources (EVERSHEIM ET AL. 1997).

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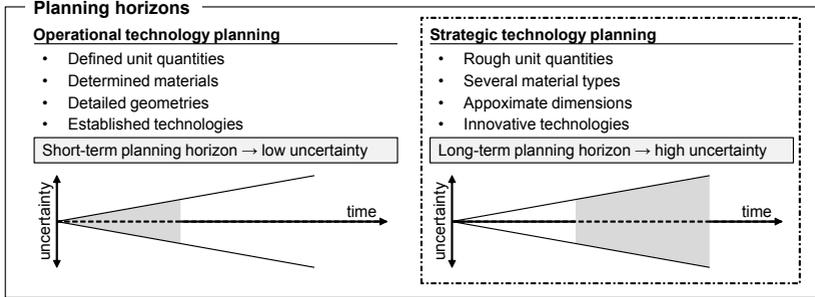


Figure 1.3: Schematic illustration of the different planning horizons with regard to operational and strategic technology planning. Adapted from REINHART ET AL. (2011) and EVERSHEIM ET AL. (1997).

In summary, current knowledge on ASSBs comes mainly from a laboratory scale. In particular, an overview of possible production scenarios and the respective challenges does not exist so far. Overall, ASSBs have been insufficiently regarded from a production point of view.

## 1.3 Research Scope

Based on the problem statement outlined in the previous section, the objective of this thesis and the research framework will be deduced in the following.

### 1.3.1 Objective

The overall aim of this thesis is to generate knowledge at the interface of product development and strategic technology planning for large-format ASSBs in order to facilitate a Simultaneous Engineering<sup>10</sup> approach (RIBBENS 2000). Hence, the **research scope** can be summarized as follows:

**The objective of this thesis is to assist stakeholders in the scale-up of production processes for large-format all-solid-state batteries.**

<sup>10</sup> *Simultaneous Engineering*, “[...] at its most rudimentary level, brings product engineering and manufacturing together” (RIBBENS 2000, p. v).

This will be guided by a methodological approach for identification and evaluation of technologies and technology chains for ASSB production. In order to support this research scope, the thesis is guided by the following **research questions**:

- **Research Question Q 1:** What are requirements for industrial fabrication of all-solid-state batteries?
- **Research Question Q 2:** Which technologies and technology chains are suitable for the production of all-solid-state batteries?
- **Research Question Q 3:** Which factors are critical<sup>11</sup> in the production of all-solid-state batteries?

By generating knowledge in the pursuit to provide answers to these questions, this thesis can help researchers in material and cell development to consider production related issues and assist in the scale-up from laboratory to pilot plant. Furthermore, the results of this thesis support industrial companies, such as the chemical industry, (potential) battery cell and component manufacturers, machine and systems engineering, and OEMs to make profound decisions within strategic technology planning.

### 1.3.2 Framework

Based on the previous considerations, the **conceptual framework** of this thesis is composed of the following three major subject areas:

- all-solid-state batteries,
- lithium-ion battery cell production, and
- strategic technology planning.

From a product perspective, the automotive sector is expected to be by far the dominating market for LICs in the upcoming decades (THIELMANN 2017). While ASSBs based on thin-film<sup>12</sup> technologies have been widely established,

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<sup>11</sup> The term *critical*, in this context, refers to the risk associated with possible technical or economical issues (WILDEMANN 2004).

<sup>12</sup> Typically, these cells are less than 15  $\mu\text{m}$  thick (DUDNEY & NEUDECKER 1999).

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e.g. in the medical sector (HOOPER & TOFIELD 1984), their limited performance<sup>13</sup> will hinder an application as automotive traction battery (JANEK & ADELHELM 2018, p. 203). The only large-format solid-state batteries (SSBs) used as traction batteries in automotive applications so far are based on polymer SEs<sup>14</sup> (MOTAVALLI 2015). However, these need to be permanently heated due to the low ionic conductivity of the polymer SE at temperatures below 60 °C (GRAPE 2015, 22ff.), limiting their practical utilization to niche applications such as Bolloré’s car sharing concept (MOTAVALLI 2015). Hence, the focus of this thesis will be on large-format lithium-based ASSBs with inorganic SEs<sup>15</sup> which will potentially be able to fulfill the requirements for automotive traction batteries, especially in battery electric vehicles (BEVs) (ANDRE ET AL. 2015; ANDRE ET AL. 2017). Furthermore, the reference frame of this thesis is narrowed down to the typical core competencies of a LIC manufacturer, i.e. the processes from electrode materials mixing to the packaging and final quality check of the battery cell (cf. figure 1.2). Thus, neither will materials synthesis be considered in detail, nor will the fabrication of components usually bought from external suppliers (such as current collectors, collector tabs, pouch foils, or housings). Only exception is the SES layer which, in contrast to the separators in conventional LIC production, will most likely need to be manufactured “in-house” by the battery cell manufacturer. Final product is the ASSB cell; module and pack integration will not be regarded. As illustrated in section 1.2, the high uncertainty associated with ASSBs limits the regarded time frame to a long-term planning horizon. Hence, the focus of this thesis lies within the activities of strategic technology management with a particular focus on strategic planning of technology chains (cf. REINHART ET AL. (2011)).

### 1.4 Research Methodology and Synopsis

Following the considerations on the research scope, the research methodology applied within this thesis will be explained and the structure of this thesis will be outlined.

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<sup>13</sup> For an overview of battery performance measures, please refer to section 2.1.

<sup>14</sup> For a classification of SE materials, please refer to section 2.2.

<sup>15</sup> Mainly sulfide- and oxide-based SEs will be regarded, cf. section 2.2.

### 1.4.1 Research Methodology

Based on the conceptual framework described in the previous section, this thesis can be categorized at the interface between fundamental and applied sciences (cf. ULRICH & HILL (1976)): While many phenomena associated with the ASSB technology are still being investigated on a basic research level, the major goal of this thesis, i.e. the transfer to an industrial scale in the frame of strategic technology planning, calls for an applied research methodology. Due to its wide applicability in engineering research, the iterative **Design Research Methodology (DRM)**<sup>16</sup> introduced by BLESSING & CHAKRABARTI (2009) has been utilized as a guidance for this thesis: Within the **Review-based Research Clarification** (cf. BLESSING & CHAKRABARTI (2009, 43ff.)), the objective, research questions, and framework of this thesis were defined. Subsequently, a **Comprehensive Descriptive Study I**<sup>17</sup> was undertaken to gather knowledge on the issues and possible solutions for industrial fabrication of ASSBs: Since, at the beginning of this research project (2015/2016), only little had been published on scalable approaches for fabricating ASSB components (JUNG ET AL. 2015; YAO ET AL. 2016), not only was a literature review undertaken on ASSBs (cf. chapter 2), but also a series of expert workshops were held<sup>18</sup>. This empirical study served to broaden the spectrum and identify possible ASSB cell designs and fabrication routes, as well as their respective challenges. As suggested by BLESSING & CHAKRABARTI (2009, 80f.), hypotheses were formulated to extend the research questions defined in the previous section. Due to the large number of recent publications (2016-2019) in the research field (cf. chapter 2), the initial

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<sup>16</sup> The DRM was introduced by BLESSING & CHAKRABARTI (2009) in order to “[support] engineering and industrial design research” (p. 2), where design “is the process through which one identifies a need, and develops a solution – a product – to fulfill this need” (p. 12) and design research “integrates [...] the development of *understanding* and the development of *support*” (p. 5) with the aim “to make design more effective and efficient in order to enable design practice to develop more successful products” (p. 12). For a classification of research types within the DRM framework, please refer to BLESSING & CHAKRABARTI (2009, 15ff., 60ff.).

<sup>17</sup> A *Comprehensive Descriptive Study I*, in contrast to a solely *Review-based Descriptive Study I*, “[...] involves a literature review as well as one or more empirical studies. The empirical studies take place when the literature review shows a lack of understanding about the chosen topic” (BLESSING & CHAKRABARTI 2009, p. 80).

<sup>18</sup> The workshops took place within the research project *Festelektrolyte als Enabler für Lithium-Zellen in automobilen Anwendungen* (Engl. *Solid electrolytes as enabler for lithium cells in automotive applications*), funded by the Federal Ministry of Education and Research (Bundesministerium für Bildung und Forschung), grant number 03XP00261 (FELIZIA). The procedure and results of these expert workshops were presented in SCHNELL ET AL. (2018).

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literature review was continuously supplemented and refined. Furthermore, the state of the art in strategic planning of technology chains was assessed with regards to the requirements resulting from the current challenges on ASSB production (cf. chapter 3). The main results of this thesis were achieved in the frame of a **Comprehensive Prescriptive Study**<sup>19</sup>, in which a methodology for strategic planning of technology chains for ASSB production was developed (cf. chapter 4). The methodology was deduced from methods applied within strategic planning of technology chains and specifically adapted towards the requirements for ASSB production, as illustrated in figure 1.4. The results were gathered and presented within the following four peer-reviewed scientific publications<sup>20</sup>, which form the core of this publication-based dissertation:

- **Publication P 1** (SCHNELL ET AL. 2017): Evaluation of technology chains for the production of all-solid-state batteries.
- **Publication P 2** (SCHNELL ET AL. 2018): All-solid-state lithium-ion and lithium metal batteries – paving the way to large-scale production.
- **Publication P 3** (SCHNELL ET AL. 2019b): Prospects of production technologies and manufacturing costs of oxide-based all-solid-state lithium batteries.
- **Publication P 4** (SCHNELL ET AL. 2020): Solid versus Liquid—A Bottom-Up Calculation Model to Analyze the Manufacturing Cost of Future High-Energy Batteries.

Finally, a case study with an international battery cell manufacturer served to evaluate the developed methodology and critically reflect its applicability in an industrial context. Therefore, an **Initial Descriptive Study II**<sup>21</sup> was undertaken,

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<sup>19</sup> A *Comprehensive Prescriptive Study*, in contrast to an *Initial* or a *Review-based Prescriptive Study*, “results in a support that is realised to such an extent that its core functionality can be evaluated for its potential to fulfil the purpose for which it was developed” (BLESSING & CHAKRABARTI 2009, p. 144).

<sup>20</sup> The full bibliography and the individual contributions of the author are listed in the appendix.

<sup>21</sup> According to BLESSING & CHAKRABARTI (2009, p. 195), an *Initial Descriptive Study II* “is required to round off a research project, that focused on the development of support to be able to draw any conclusions about the relation between the support and the aims of the research project. [...] Minimally required are an indication of the applicability, usability and usefulness of the support; an indication of the issues, factors and links that need detailed evaluation; a suggestion for a proper Evaluation Plan.” In contrast, a *Comprehensive Descriptive Study II* is typically employed to “follow up another research project in which design support has been developed” (BLESSING & CHAKRABARTI 2009, p. 195). In the underlying case, such a study should involve a validation using at least pilot scale equipment, for which the outcomes of this thesis lay a sound foundation.

which was further used to assess the requirements fulfillment, to discuss the earlier defined hypotheses, and to conclude on future research (cf. chapters 5 and 6).

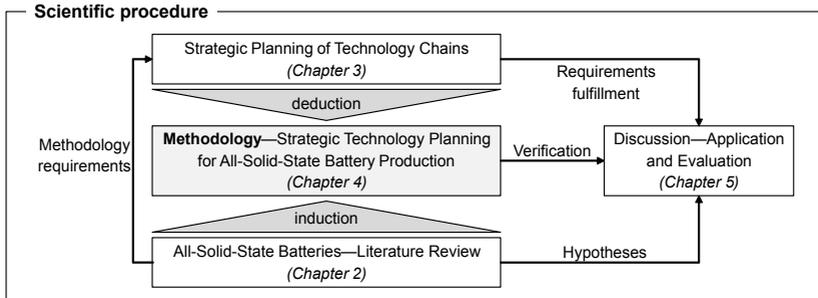


Figure 1.4: Procedure applied to develop and assess the methodology for strategic technology planning for ASSB production within the underlying framework.

### 1.4.2 Structure of the Thesis

As outlined above, this thesis is a publication-based dissertation. Following the considerations within the previous subsection on the DRM by BLESSING & CHAKRABARTI (2009) and the research methodology for applied sciences by ULRICH & HILL (1976), the remainder of thesis is structured as follows<sup>22</sup> (figure 1.5): Based on the objective, research questions, and framework formulated within the underlying chapter, a literature review on the current state of research and respective manufacturing approaches for ASSBs is given in **chapter 2**. Within the course of this literature review, hypotheses are formulated to refine the earlier defined research questions (BLESSING & CHAKRABARTI 2009, 80f.), and challenges to be addressed within this thesis are summarized. Hence, requirements on a methodology for strategic planning of ASSB production are deduced in **chapter 3**, which serve as a basis to critically assess the state of the art in strategic planning of technology chains and to derive the resulting action required. **Chapter 4** outlines the methodology which was adapted from existing technology planning approaches towards the specific requirements

<sup>22</sup> Similar considerations can be found, e.g. in HEIMES (2014, 4ff.), and PLEHN (2018, 16ff.).

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for strategic planning of ASSB production: Based on the general procedure developed in **P 1**, the results of an empirical study are illustrated in **P 2** to gather requirements and challenges for industrial fabrication of ASSBs and to identify possible processing routes. In **P 3**, technologies applied in the fabrication of ceramic energy storage and conversion systems are evaluated with regards to their applicability in ASSB production, followed by a top-down calculation of the expected manufacturing cost. In order to refine these computations and to compare different scenarios for ASSB production, a bottom-up calculation model is presented in **P 4**. Based on these findings, results of an application of the methodology with an international battery cell manufacturer are presented in **chapter 5**. This serves as an input to evaluate and critically reflect the outcomes of this thesis with regards to technical and economic aspects and to discuss the hypotheses defined earlier. Finally, **chapter 6** gives a summary of the thesis and concludes with an outlook on future research.

Structure of the thesis		
<b>Chapter 1</b>	<b>Introduction</b> <ul style="list-style-type: none"> <li>Objective &amp; research questions</li> <li>Framework</li> </ul>	<i>Research Clarification (Review-based)</i>
<b>Chapter 2</b>	<b>All-Solid-State Batteries—Literature Review</b> <ul style="list-style-type: none"> <li>Literature review</li> <li>Hypotheses</li> </ul>	<i>Descriptive Study I (Comprehensive)</i>
<b>Chapter 3</b>	<b>Strategic Planning of Technology Chains</b> <ul style="list-style-type: none"> <li>Methodology requirements</li> <li>State of the art</li> <li>Action required</li> </ul>	
<b>Chapter 4</b>	<b>Methodology—Strategic Technology Planning for All-Solid-State Battery Production</b> <ul style="list-style-type: none"> <li>Requirements for ASSB production</li> <li>Technologies &amp; technology chains</li> <li>Cost model</li> </ul> 	<i>Prescriptive Study (Comprehensive)</i>
<b>Chapter 5</b>	<b>Discussion—Application and Evaluation</b> <ul style="list-style-type: none"> <li>Application example</li> <li>Critical reflection</li> <li>Discussion of hypotheses &amp; research questions</li> </ul>	<i>Descriptive Study II (Initial)</i>
<b>Chapter 6</b>	<b>Conclusion</b> <ul style="list-style-type: none"> <li>Summary</li> <li>Further research</li> </ul>	

Figure 1.5: Overview of the chapters within this thesis and integration of the publications **P 1** to **P 4**. The types of study (in italics) correspond to the different stages according to the DRM by BLESSING & CHAKRABARTI (2009).

## 2 All-Solid-State Batteries—Literature Review

Based on the research scope and the research questions presented in the previous chapter, this chapter is dedicated to give an overview of the current state of research on all-solid-state batteries (ASSBs). Section 2.1 will provide a basic understanding of the functionality and performance measures of ASSBs. This will serve as a guideline for the analysis of materials and the respective interfaces in section 2.2. In section 2.3, an overview of laboratory manufacturing approaches for the different ASSB components and considerations on ASSB scale-up will be given, before summarizing the resulting challenges in section 2.4. Following the DRM by BLESSING & CHAKRABARTI (2009, 80f.), several hypotheses will be formulated within this chapter to refine the research questions outlined in the previous chapter. These hypotheses are to be confirmed or refuted in the course of this thesis.

### 2.1 Fundamentals

This section serves to explain the fundamental properties of ASSBs. In subsection 2.1.1, basic definitions and the functionality of an ASSB will be explained, before deducing performance measures and corresponding requirements on the ASSB components and materials in subsection 2.1.2.

#### 2.1.1 Functional Principle

A **battery** is an energy storage device which converts chemical energy into electrical energy (WINTER & BRODD 2004, p. 4252). The smallest functional unit of a battery is a **galvanic cell**: During discharge of the battery, ions are transferred from the **anode** to the **cathode** via the **electrolyte**, while the **separator** physically and electrically isolates anode and cathode from each other (LEUTHNER 2018, p. 14). Hence, in order to ensure charge neutrality, electrons are forced to flow from the anode to the cathode via an external circuit (LEUTHNER 2018, p. 14). By convention, the anode is referred to as the negative pole and the cathode as the positive pole of the battery (DOEFF 2013, p. 7). In contrast to a primary

battery, in which the discharge process takes place irreversibly, a **secondary battery** can be recharged<sup>1</sup> (LEUTHNER 2018, p. 13). Lithium based batteries can be grouped into **lithium-ion batteries (LIBs)** and **lithium metal batteries (LMBs)**: Whereas in LIBs, the lithium-ions in the anode are stored in a host material (intercalation materials, such as graphite), in LMBs, lithium is present in its metallic form (MOELLER 2018, pp. 5, 8) (cf. section 2.2). In an **ASSB**, a **solid electrolyte (SE)** is employed to replace the liquid electrolyte and the porous separator which are typically used in conventional LIBs (cf. section 1.1).

Figure 2.1 (left image) illustrates the functional principle of an ASSB with a lithium metal anode: Here, the transfer of Li-ions from anode to cathode and vice versa takes place via the **solid electrolyte separator (SES)** which enables ionic transport<sup>2</sup>, but ensures electric isolation to hinder short circuiting or self discharge of the ASSB (GOODENOUGH & KIM 2010; HAN ET AL. 2019). During discharge (right arrows in equation 2.1 and equation 2.2) of the battery, Li-ions are stripped from the metal anode



(CHENG ET AL. 2017) and intercalated into the cathode<sup>3</sup>



(LIU ET AL. 2018), with  $M$  a (combination of) transition metal(s), such as Co, Ni, or Mn. Vice versa, during charge (left arrows in equation 2.1 and equation 2.2), Li-ions are deintercalated from the cathode and plated at the anode in the form of metallic lithium. In order to ensure sufficiently fast ion transport inside the electrodes, SE particles may be inserted to form composite electrodes (HOOPER & TOFIELD 1984; SAKUDA 2018). Similarly, conductive agents in the electrodes enhance the transport of electrons towards the current collectors, from which the

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<sup>1</sup> If not otherwise denoted, only secondary batteries will be considered in this thesis.

<sup>2</sup> The transfer mechanism of ions is dependent on the SE used and will be briefly explained in section 2.2.

<sup>3</sup> Only intercalation cathode materials will be regarded in this thesis, cf. subsection 2.2.1.

electrons are drawn to provide an external current (VUORILEHTO 2018, p. 23). In order to provide cohesion inside the electrode and adhesion towards the current collectors, binders may also be integrated (not illustrated in figure 2.1) (VUORILEHTO 2018, p. 23).

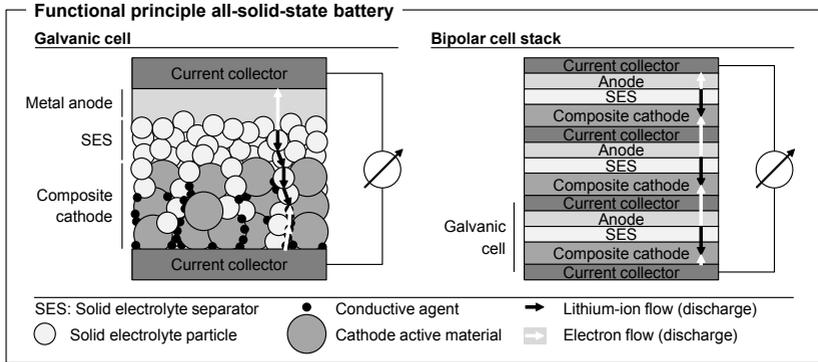


Figure 2.1: Schematic illustration of the functional principle of an all-solid-state battery for a galvanic cell (left) and a bipolar cell stack (right) during discharge. Adapted from PLACKE ET AL. (2017) and HU (2016).

The **energy**  $E$  (in Wh) stored in a battery is calculated by

$$E = C \cdot \bar{V} \quad (2.3)$$

with

- $C$  the discharge **capacity**<sup>4</sup> (in Ah) and
- $\bar{V}$  the average discharge **voltage**<sup>5</sup> (in V)

(LEUTHNER 2018, p. 16). In order to increase the energy content of a battery, multiple galvanic cells can be stacked and encased to form a **battery cell** (LEUTHNER 2018, p. 15; PETTINGER ET AL. 2018, 220f.). In a conventional lithium-ion cell (LIC), typically, the electrodes are symmetrically applied on both sides of

<sup>4</sup> The *capacity* is defined as “the amount of electric charge a power source can deliver under specific discharge conditions” (LEUTHNER 2018, p. 16).

<sup>5</sup> The *cell voltage* results from the potential difference of the positive and negative electrode (WOEHRLE 2018, p. 104).

the current collectors (BRODD & TAGAWA 2002, p. 276). This enables a parallel connection and, thus, a capacity increase of the stacked or wound galvanic cells (PETTINGER ET AL. 2018, 220f.). However, additional space is required in the cell housing to allow for joining of all anode and cathode current collectors and current collector tabs, respectively (PETTINGER ET AL. 2018, p. 217). Although this design seems to be a plausible option also for ASSBs (ITO ET AL. 2014), the spatial confinement of the SES may also enable a so-called **bipolar stacking**, as illustrated in figure 2.1, right image (HOOPER & TOFIELD 1984; NAM ET AL. 2015): Here, the anode and cathode are applied on opposite sides of the same current collector, resulting in a serial connection of the stacked cell and an increase of the overall cell voltage with every set of layers<sup>6</sup>. Since the current in a bipolar design is only drawn from the outermost layers of the cell stack, less space is required for joining of the current collector tabs, i.e. the space in the packaging can be used more efficiently (SATOU 2017). Furthermore, smaller currents are required due to the higher operating voltage.

### 2.1.2 Performance Measures and Requirements

Main performance measures of a LIC for automotive applications are the specific energy and energy density, the power, charging current, and low temperature performance, as well as lifetime, safety, and cost<sup>7</sup> (ANDRE ET AL. 2015). These requirements may be contradictory to each other, calling for very careful tuning of the material properties and cell design. The implications of these performance measures on the component and materials of an ASSB will be explained in the following paragraphs.

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<sup>6</sup> In this concept, a bipolar current collector is required, enabling electron conduction but blocking ion transfer. The bipolar current collector must be electrochemically stable against both the anode and cathode potential and mechanically stable to prevent short circuiting (TANAKA 2016). In a conventional LIC, the liquid electrolyte interconnects the whole battery, which would lead to an ionic short circuit (around the current collector) for the serial connection (KATO ET AL. 2012). Nonetheless, concepts for confining the liquid electrolyte in different compartments to enable bipolar stacking of conventional LIC have been suggested in literature (e.g. by MARSH ET AL. (1997)).

<sup>7</sup> Although not elaborated in detail, also the environmental friendliness, social impact, and sustainability can play an important role, especially in materials selection (ANDRE ET AL. 2017).

## Specific Energy and Energy Density

The **specific energy**  $E_{\text{spec}}$  (typically given in Wh/kg) and the **energy density**  $E_{\text{vol}}$  (in Wh/L) of a battery are calculated by

$$E_{\text{spec}} = c_{\text{spec,c}} \cdot \bar{V} \cdot \frac{M_c}{\sum M_i} \quad (2.4)$$

and

$$E_{\text{vol}} = c_{\text{spec,c}} \cdot \bar{V} \cdot \frac{M_c}{\sum M_i / \rho_i}, \quad (2.5)$$

respectively (MCCLOSKEY 2015). Here,

- $c_{\text{spec,c}}$  is the cathode active material specific capacity<sup>8</sup> (in mAh/g),
- $M_c$  the cathode active material areal loading (in mg/cm<sup>2</sup>),
- $M_i$  the grammage<sup>9</sup> (or loading) of the individual cell components (in mg/cm<sup>2</sup>), and
- $\rho_i$  the density of the individual cell components (in g/cm<sup>3</sup>).

As easily deduced from these equations, the introduction of inactive materials or void spaces (e.g. porosities) on the component, cell, and battery pack level reduces the overall specific energy and energy density compared to the material level (ANDRE ET AL. 2015; BETZ ET AL. 2019). For instance, if a specific energy of 250 Wh/kg on the battery pack level is targeted for BEVs in 2025, at least 300 Wh/kg will be required on the battery cell level to account for additional weight of the cooling system, battery management system, etc. (ANDRE ET AL. 2017). Similarly, an energy density target of 800 Wh/L on cell level by 2025 can be derived, which is of major interest especially in the automotive industry where the space for battery integration is limited (ANDRE ET AL. 2015).

In order to achieve the targeted specific energies and energy densities on cell level, the following conclusions can be drawn for the ASSB components and materials (SES, cathode, anode): For the SE, as an inactive component, the more material used, the lower the resulting energy density and specific energy. Hence,

<sup>8</sup> As outlined in section 2.2, the focus of this thesis will be on lithiated cathode materials, i.e. the cathode active material (cf. section 2.2) provides the Li-ions used for cycling the battery.

<sup>9</sup> The cited literature uses the term "weight". However, technically,  $M_i$  corresponds to an areal density. On the cell level, it may be more convenient to replace  $M_i$  by the total masses of the individual components (in g) with respect to the cathode lateral area (in cm<sup>2</sup>).

## 2 All-Solid-State Batteries—Literature Review

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from an energy perspective, a SES layer as thin as possible is to be targeted and the content of SE material in the electrodes should be minimized. A low density SE material is to be preferred to ensure high specific energy (WU ET AL. 2016). Based on geometrical considerations (PETRI ET AL. 2015), the electrode thicknesses  $d_{\text{el}}$  (in  $\mu\text{m}$ ) can be calculated as

$$d_{\text{el}} = \frac{M_{\text{a,c}}/\rho_{\text{a,c}} + \sum M_{\text{p}}/\rho_{\text{p}}}{(1 - x_{\text{p}})}, \quad (2.6)$$

with

- $M_{\text{a,c}}$  the anode/cathode active material areal loading (in  $\text{mg}/\text{cm}^2$ ),
- $\rho_{\text{a,c}}$  the anode/cathode active material crystallographic density (in  $\text{g}/\text{cm}^3$ ),
- $M_{\text{p}}$  the grammage (or loading) of the passive electrode components (in  $\text{mg}/\text{cm}^2$ ),
- $\rho_{\text{p}}$  the crystallographic density of the passive electrode components (in  $\text{g}/\text{cm}^3$ ), and
- $x_{\text{p}}$  the electrode porosity (in vol%).

Rearranging leads to

$$M_{\text{a,c}} = \rho_{\text{a,c}}[d_{\text{el}}(1 - x_{\text{p}}) - \sum M_{\text{p}}/\rho_{\text{p}}] \quad (2.7)$$

which, inserted into equation 2.4, reveals that a thick and dense cathode layer with high active material content is desirable to achieve a high specific energy (WU ET AL. 2016; SAKUDA 2018). However, the obtainable thickness and active material content are typically limited by ion or electron diffusion (cf. next paragraph). For the cathode active material, next to a high specific capacity and high potential (compared to a reference anode material, e.g. lithium), also a high density is required. The areal loading and, thus, the thickness of the anode layer should be balanced to match the area specific capacity

$$C_{\text{area}} = c_{\text{spec,c}} \cdot M_{\text{c}} \quad (2.8)$$

(in  $\text{mAh}/\text{cm}^2$ ) of the cathode (SCHÜNEMANN 2016). Typically, for lithiated cathode materials, the anode is overbalanced to ensure that all lithium drawn from the cathode during charge can be stored (for intercalation materials (EBERMAN ET AL. 2010)) or to account for irreversible losses during cycling (for lithium

metal (GENOVESE ET AL. 2018), cf. section 2.2). For the anode active material, a high specific capacity is desired, as well as a low potential (given a reference cathode material) to ensure a high operating voltage (ANDRE ET AL. 2017).

### Power, Charging Current, Low Temperature Power

Similar, yet partly contradictory conclusions can be drawn for the **power, charging current** and **low temperature power**, i.e. the energy which can be drawn from (or inserted into) the battery in a specific amount of time under certain conditions (ANDRE ET AL. 2015). These parameters are mainly governed by the **current density**  $J$  (in mA/cm<sup>2</sup>), as will be explained in the following: Based on Ohm's law, the voltage drop  $\Delta V$  (in V) across the SES layer can be estimated as

$$\Delta V = I \cdot R = I \cdot \frac{d_s}{\sigma_i \cdot A} = \frac{J \cdot d_s}{\sigma_i} \quad (2.9)$$

(WU ET AL. 2016). Here,

- $I$  is the current (in A),
- $R$  the Ohmic resistance (in  $\Omega$ ),
- $d_s$  the layer thickness (in  $\mu\text{m}$ ),
- $\sigma_i$  the ionic conductivity (in S/cm), and
- $A$  the lateral area (in cm<sup>2</sup>).

The higher this voltage drop, the lower the operating voltage during discharge or the higher the overpotential which needs to be applied during charge of the battery. Therefore, especially for high current densities, a thin SES layer and a high total ionic conductivity<sup>10</sup> ( $>10^{-4}$  S/cm) over a wide temperature range for the SE are required (GOODENOUGH & KIM 2010). Similar considerations can be made for the electrodes, where ionic and electric pathways are required: Here, a tradeoff between power and energy is required concerning the electrode thickness as well as the share of SE materials and conductive agents (SAKUDA 2018). Additionally, interface engineering is required to reduce the transfer resistance between the particles (cf. section 2.2) (LUNTZ ET AL. 2015). On the material level, a high rate capability of the active materials is required, i.e. the

<sup>10</sup> The *total ionic conductivity* takes into account the bulk conductivity (inside the SE material) and the grain boundary resistance (ZHANG ET AL. 2018).

capability to store and release Li-ions and electrons in a short time interval (ANDRE ET AL. 2015).

### **Lifetime, Safety, Cost**

Other important requirements are the calendaric and cyclic **lifetime**, as well as **safety** and **cost** (ANDRE ET AL. 2015). In order to ensure sufficient mechanical stability, a certain thickness of the SES is required, especially with regard to volume changes of the active materials during cycling (KERMAN ET AL. 2017). Additionally, when employing a lithium metal anode, the resulting layer must be impenetrable to lithium dendrites (cf. section 2.2) (TIKEKAR ET AL. 2016). For the SE materials, in order to achieve a long lifetime during use of the ASSB, a high mechanical strength and thermal stability, as well as electrochemical stability towards anode and cathode materials are required (GOODENOUGH & KIM 2010). A negligible electronic conductivity of the SE ( $<10^{-10}$  S/cm) hinders short circuiting and self discharge of the ASSB (GOODENOUGH & KIM 2010; HAN ET AL. 2019). For the electrodes, binders can be integrated to efficiently buffer volume changes and ensure mechanical integrity (KERMAN ET AL. 2017). In order to reduce degradation effects induced by mechanical strain, active materials with low volume change during cycling are advantageous for long cycle life (cf. SAKUDA (2018) and references therein). Last but not least, cost plays an important role for mass commercialization: Targets on battery pack level for the automotive industry are below 150 \$/kWh. As illustrated by SCHMUCH ET AL. (2018), the material cost for SE materials, but also for lithium metal, have a significant influence on the overall ASSB cost. Up to date, these costs are accompanied by a high uncertainty. Since the material costs are usually given in \$/kg, a higher SE density typically leads to higher cost (SCHMUCH ET AL. 2018).

These partly conflicting requirements call for elaborate cell design. This has implications on the materials, as will be outlined in the following sections.

### **2.2 Materials and Interfaces**

Based on the requirements deduced in the last section, a brief overview of cathode (subsection 2.2.1), anode (subsection 2.2.2), and SE (subsection 2.2.3)

materials will be given. Furthermore, challenges at the respective interfaces will be summarized in subsection 2.2.4.

### 2.2.1 Cathode Materials

Typical cathode active materials for LIBs comprise phosphates, such as  $\text{LiFePO}_4$  (LFP), and oxides, such as  $\text{LiCoO}_2$  (LCO) and  $\text{LiMn}_2\text{O}_4$  (LMO) (GRAF 2018). However, LFP and LMO suffer from relatively low specific energies, while the high Co content in LCO leads to environmental, social, and cost concerns. Among the most promising candidates for future automotive applications are  $\text{Li}(\text{Ni}_{1-x-y}\text{Co}_x\text{Al}_y)\text{O}_2$  (NCA) and  $\text{Li}(\text{Ni}_{1-x-y}\text{Mn}_x\text{Co}_y)\text{O}_2$  (NMC) with reduced Co-content (ANDRE ET AL. 2015). This entails in particular the Ni-rich  $\text{Li}(\text{Ni}_{0.8}\text{Mn}_{0.1}\text{Co}_{0.1})\text{O}_2$  (NMC811) and Li-rich  $x \text{Li}_2\text{MnO}_3 \cdot (1-x) \text{LiMO}_2$  (HE-NMC) with  $M = \text{Co}, \text{Ni}, \text{Mn}$  (THACKERAY ET AL. 2007; QIU ET AL. 2016). These materials show a comparatively high level of development and capability to satisfactorily meet the above described requirements. Further materials possibly to be considered are high voltage spinel  $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$  (LNMO) and conversion cathodes, such as  $\text{FeF}_3$  (ANDRE ET AL. 2015). The latter, however, show a very low degree of maturity so far and will therefore not be included in the further considerations. Table 2.1 gives an overview of the discussed materials with typical data for specific energy calculations.

Table 2.1: Overview of the discussed cathode active materials with typical values for the specific capacity  $c_{\text{spec},c}$  and the operating voltage  $V$ . Adapted from ANDRE ET AL. (2015), QIU ET AL. (2016), and GRAF (2018).

Material	$c_{\text{spec},c}$ in mAh/g	$\bar{V}$ (vs. Li/Li <sup>+</sup> ) in V
LMO	100	4.1
LFP	160	3.4
LCO	160	3.9
LNMO	137	4.7
NCA	200	3.7
NMC811	200	3.7
HE-NMC	300	3.4

### 2.2.2 Anode Materials

Conventional LIC anodes are typically based on graphite or  $\text{Li}_4\text{Ti}_5\text{O}_{12}$  (LTO) (WURM ET AL. 2018). Both materials have also been successfully employed in composite electrodes for ASSBs, e.g. to achieve high power (up to  $12 \text{ mA}/\text{cm}^2$ ) or high area specific capacities (up to  $15.7 \text{ mAh}/\text{cm}^2$ ) (KATO ET AL. 2016c; KATO ET AL. 2018). In order to enable higher energy densities, anodes based on silicon and metallic lithium seem promising candidates<sup>11</sup> (ANDRE ET AL. 2017), although integration into large-format ASSB has been challenging due to the large volumetric change during cycling (BAGGETTO ET AL. 2008; VARZI ET AL. 2016). Due to the high specific capacity and low operating voltage (cf. table 2.2), the focus of this thesis will be on lithium metal as anode material. Nonetheless, many of the considerations made for composite cathodes (cf. subsection 2.3.3) could also be applied to composite anodes using, for instance, graphite or silicon-based anodes.

Table 2.2: Overview of the discussed anode active materials with typical values for the specific capacity  $c_{\text{spec},a}$  and the operating voltage  $V$ . Based on WURM ET AL. (2018).

Material	$c_{\text{spec},a}$ in mAh/g	$\bar{V}$ (vs. Li/Li <sup>+</sup> ) in V
LTO	160	1.5
graphite	372	0.1
silicon	4212	0.4
lithium	3860	0

### 2.2.3 Solid Electrolyte Materials

Different approaches to classify SE materials can be found in literature. SEs are typically grouped into organic and inorganic materials (cf. figure 2.2). The first mainly include solid polymer and gel polymer<sup>12</sup> SEs. The latter typically comprise sulfide- and oxide-based SEs. To combine the properties of organic

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<sup>11</sup> Phosphate-based anodes, as also suggested by ANDRE ET AL. (2017), display a very low degree of maturity (ibid.) and will not be considered in this thesis.

<sup>12</sup> Gel-polymer solid electrolytes typically comprise liquid components (KELLER ET AL. 2018) and will, therefore, not be considered in this thesis.

and inorganic SEs, hybrid SEs have also been suggested. In the following subsections, a brief overview of both polymer and inorganic SE materials will be given.

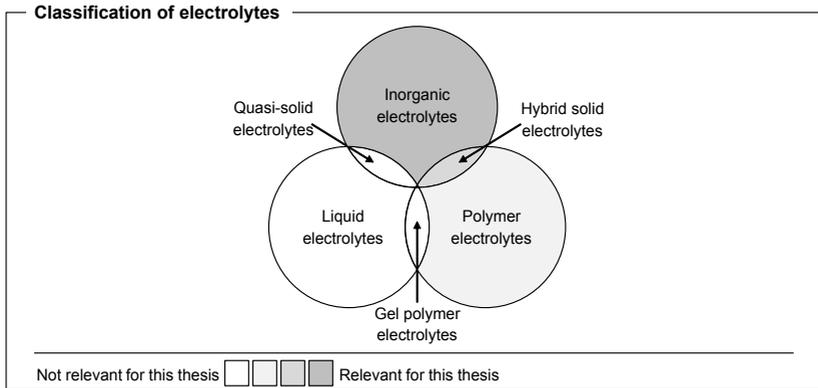


Figure 2.2: Classification of different types and combinations of electrolytes. Adapted from KELLER ET AL. (2018).

### Polymer-based solid electrolytes

In polymer SEs, ions are complexed by the polar groups of the polymer chains and ion transfer typically occurs above the glass transition temperature, assisted by segmental motion of the polymer chains (BERTHIER ET AL. 1983; MEYER 1998). Among the most widely studied polymer systems for SEs is poly(ethylene oxide) (PEO), combined with a lithium salt such as LiTFSI or LiPF<sub>6</sub> (YUE ET AL. 2016). Since they form stable passivating interfaces with lithium metal, polymer SEs have been successfully commercialized in lithium metal secondary batteries, for instance as traction batteries in electric vehicles (MOTAVALLI 2015). Due to their elastic and plastic behavior, polymer SEs can compensate for volume changes of the active materials during cycling and are also relatively simple to process (GRAPE (2015), cf. section 2.3). However, their low ionic conductivity at room temperature (typically  $10^{-6}$  S/cm to  $10^{-5}$  S/cm) makes permanent heating in the range of 60 °C to 80 °C necessary (MOTAVALLI 2015). Furthermore, most polymer-based SEs show limited stability against cathode potentials larger than 4 V, narrowing down the selection of suitable cathode materials to lower

voltage materials (such as LFP, cf. subsection 2.2.1) (YUE ET AL. 2016). Hence, the use of polymer based SE has been limited to applications where permanent heating and limited energy density can be accepted (such as Bollore's car sharing concept)<sup>13</sup>. Due to their stability against lithium anodes, thin ( $\approx 1 \mu\text{m}$ , cf. equation 2.9) polymer SE layers could also be applied as protective layers in combination with inorganic SEs (ATES ET AL. 2019).

### Inorganic solid electrolytes

In inorganic SEs, ions travel via crystalline lattice sites, for example by "hopping" to adjacent vacant sites such as lattice defects (BACHMAN ET AL. 2016). In contrast to liquid and polymer based electrolytes, inorganic SEs are single-ion conductors, i.e. Li-ions are the only mobile species (ZHANG ET AL. 2018). Therefore, no concentration gradients are observed in inorganic SEs, potentially enabling higher current densities (KATO ET AL. 2016c; KATO ET AL. 2018). Depending on their structure, inorganic SEs can be classified as crystalline, glass-ceramic, or amorphous (VARZI ET AL. 2016). They are typically grouped into oxide- and sulfide-based SEs<sup>14</sup> (VARZI ET AL. 2016):

Among the **oxide-based SEs**, lithium phosphorous oxynitride  $\text{Li}_x\text{PO}_y\text{N}_z$  (LIPON) has been widely established for use in thin-film ASSBs (REN ET AL. 2015b; FAN ET AL. 2018). As an amorphous ion conductor it is often used in the composition  $\text{Li}_{2.88}\text{PO}_{3.73}\text{N}_{0.14}$ . Although theoretical calculations reveal a limited thermodynamic stability, LIPON forms stable interfaces with metallic lithium (ZHU ET AL. 2016). The room temperature ionic conductivity is in the order of  $10^{-6} \text{ S/cm}$  (BATES ET AL. 1993), which is not sufficient for large-format ASSBs—especially in automotive applications. In contrast, the phosphate based  $\text{LiM}_2(\text{PO}_4)_3$  family<sup>15</sup> ( $M = \text{Ti, Ge, and others, cf. ZHANG ET AL. (2018) and refer-$

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<sup>13</sup> To increase the room temperature conductivity, liquid plasticizers such as propylene carbonate or ethylene carbonate are often added (YUE ET AL. 2016). However, as the focus of this thesis is on ASSBs, these semi-liquid electrolytes will not be further considered. Recent research efforts indicate that acceptable ionic conductivities above  $10^{-4} \text{ S/cm}$  at room temperature could be achievable, e.g. for poly(propylene carbonate)-based polymer SEs (ZHANG ET AL. 2015).

<sup>14</sup> Oxide-based SEs, in this context, also comprise oxynitrides and phosphates, while sulfide-based SEs also include thiophosphates. Further SE materials, such as halide- and hydride-based SEs, will not be considered in this thesis.

<sup>15</sup> Typically, these materials are classified as NAtrium SuperIonic CONductors (NASICON) because of their crystal-structure, which was first identified for sodium-ion conductors (GOODENOUGH ET AL. 1976; ZHANG ET AL. 2018).

ences therein), shows ionic conductivities up to  $5 \times 10^{-3}$  S/cm (THOKCHOM & KUMAR 2008; REN ET AL. 2015b). While the high Ge price realistically excludes SE systems such as  $\text{Li}_{1+x}\text{Al}_x\text{Ge}_{2-x}(\text{PO}_4)_3$  (LAGP) (ZHANG ET AL. 2012), especially  $\text{Li}_{1+x}\text{Al}_x\text{Ti}_{2-x}(\text{PO}_4)_3$  (LATP) seems to be promising: The crystalline phase has a total ionic conductivity of  $7 \times 10^{-4}$  S/cm at room temperature (AONO 1989; AONO 1990), and the glass-ceramic analogue, prepared by elaborate heat treatment (up to  $950^\circ\text{C}$ ), exhibits conductivities as high as  $1.3 \times 10^{-3}$  S/cm (FU 1997). LATP seems to be chemically stable in ambient conditions (FAN ET AL. 2018). Major disadvantage is the reduction of  $\text{Ti}^{4+}$  ions when in contact with Li metal or graphite anodes, leading to electron conducting phases which easily result in short circuiting of the ASSB (HARTMANN ET AL. 2013).

Next to oxynitrides and phosphates, a large variety of oxides with different crystal structures, such as perovskites and garnets<sup>16</sup>, has been investigated as Li-ion conductors (ZHANG ET AL. 2018). The perovskite structured lithium lanthanum titanate (e.g.  $\text{Li}_{0.5}\text{La}_{0.5}\text{TiO}_3$  (LLTO)) has been intensively studied due to its bulk ionic conductivity in the range of  $10^{-3}$  S/cm (INAGUMA ET AL. 1993). However, the large grain boundary resistance results in total conductivities which are two orders of magnitude lower (ITOH ET AL. 1994). Furthermore, similar to LATP, the reduction of  $\text{Ti}^{4+}$  ions at low anode potentials leads to an undesired electrical conductivity (BIRKE 1997). Due to their wide electrochemical stability window, garnets with the typical composition  $\text{Li}_x\text{La}_y\text{Zr}_z\text{O}_{12}$  (LLZ) are currently among the most promising oxide-based SE materials. In contrast to most other SE materials, LLZ seems electrochemically stable against metallic lithium (ZHU ET AL. 2016), although reduction at grain boundaries has been observed (REN ET AL. 2015a).  $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$  (LLZO) exerts an ionic conductivity of  $3 \times 10^{-4}$  S/cm (MURUGAN ET AL. 2007). Higher conductivities in the order of  $10^{-3}$  S/cm could be obtained, e.g. for  $\text{Li}_{6.4}\text{La}_3\text{Zr}_{1.4}\text{Ta}_{0.6}\text{O}_{12}$  (LLZ:Ta) (LI ET AL. 2012) or  $\text{Li}_{6.5}\text{La}_3\text{Zr}_{1.75}\text{Te}_{0.25}\text{O}_{12}$  (LLZ:Te) (DEVIANNAPOORANI ET AL. 2013). Sintering temperatures in the order of  $1000^\circ\text{C}$  or higher are required to obtain dense samples with ionic conductivity above  $10^{-3}$  S/cm (LI ET AL. 2012). Although previously reported to be stable in ambient conditions, LLZ has been shown to degrade in humidity and carbon dioxide ( $\text{CO}_2$ ), leading to

<sup>16</sup> Further oxides comprise antiperovskites or so-called Lithium Superionic CONductors (LISICON) (ZHANG ET AL. 2018).

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the formation of undesirable carbonate phases on the surface (JIN & MCGINN 2013b). The high density of LLZ ( $5.1 \text{ g/cm}^3$ ) is a disadvantage with regard to the specific energy (PLACKE ET AL. 2017).

Most oxide-based SEs typically form hard crystallites, resulting in a rather brittle nature and requiring high temperature processing to overcome grain boundary resistances (SAKUDA ET AL. 2013; REN ET AL. 2015b). In addition to the high energy consumption (TROY ET AL. 2016), the sintering step also results in technical challenges, such as unwanted side reactions with cathode materials (cf. subsection 2.3.3). With regards to the research questions formulated in subsection 1.3.1, the following hypothesis will be formulated:

**Hypothesis H 1: For oxide-based ASSBs, sintering is a critical process step from a technical and economic perspective.**

In **sulfide-based SEs**, the Li-ions can move more freely due to the lower binding energy between sulfur- and Li-ions, resulting in high ionic conductivities and low grain boundary resistances (CHEN ET AL. 2018b). In contrast to their oxide-based counterparts, sulfides typically have a higher plasticity, leading to a rather ductile nature (SAKUDA ET AL. 2013). Among the promising sulfide-based SEs is the glass-ceramic  $\text{Li}_2\text{S}-\text{P}_2\text{S}_5$  (LPS) system, which can consist of amorphous and crystalline phases: Glass-ceramic  $\text{Li}_2\text{S}-\text{P}_2\text{S}_5-\text{LiI}$  (LPSI) has shown ionic conductivities in the range of  $10^{-3} \text{ S/cm}$  (MERCIER ET AL. 1981). The crystalline  $\text{Li}_6\text{PS}_5\text{X}$  (LPSX) ( $X = \text{Cl, Br, I}$ , (DEISEROTH ET AL. 2008)) family<sup>17</sup> has shown ionic conductivities up to  $1.3 \times 10^{-3} \text{ S/cm}$ , e.g. for  $\text{Li}_6\text{PS}_5\text{Cl}$  (LPSCI) (BOULIN-EAU ET AL. 2012). When heating  $70 \text{ Li}_2\text{S} \cdot 30 \text{ P}_2\text{S}_5$  above the glass transition temperature ( $230 \text{ }^\circ\text{C}$  to  $280 \text{ }^\circ\text{C}$ ), the crystalline phase  $\text{Li}_7\text{P}_3\text{S}_{11}$  is formed, resulting in ionic conductivities as high as  $1.7 \times 10^{-2} \text{ S/cm}$  (SEINO ET AL. 2014). Different compositions of  $\text{Li}_2\text{S}-\text{GeS}_2-\text{P}_2\text{S}_5$  (LGPS) have been intensively investigated due to their high ionic conductivities, up to  $2.2 \times 10^{-3} \text{ S/cm}$  for  $\text{Li}_{3.25}\text{Ge}_{0.25}\text{P}_{0.75}\text{S}_4$ <sup>18</sup> (KANNO & MASAHIRO 2001) and  $1.2 \times 10^{-2} \text{ S/cm}$  for  $\text{Li}_{10}\text{GeP}_2\text{S}_{12}$  (KAMAYA ET AL. 2011). Similar results have been achieved by replacing the expensive Ge with Si or Sn (e.g. BRON ET AL. (2013)) and aliovalent-ion doping: The highest ionic conductivities for SEs of  $2.5 \times 10^{-2} \text{ S/cm}$  reported so far have been

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<sup>17</sup> The crystalline structure is referred to as argyrodite (DEISEROTH ET AL. 2008).

<sup>18</sup> This composition is often referred to as Thio-LISICON (ZHANG ET AL. 2018).

achieved for  $\text{Li}_{9.54}\text{Si}_{1.74}\text{P}_{1.44}\text{S}_{11.7}\text{Cl}_{0.3}$  (LSiPSCI) (KATO ET AL. 2016c). However, most sulfides show limited stability towards anode and cathode potentials (ZHU ET AL. 2016), often leading to insulating phases with high resistance (WENZEL ET AL. 2016). Among the main drawbacks of most sulfide-based SEs is the high reactivity in ambient atmosphere, especially the generation of toxic ( $\text{H}_2\text{S}$ ) when in contact with humidity (MURAMATSU ET AL. 2011). This results in the following hypothesis:

**Hypothesis H 2: For sulfide-based ASSBs, the processing atmosphere is a critical influence factor from a technical and economic perspective.**

### Concluding Remarks on Solid Electrolyte Materials

An overview of the SEs discussed in the previous subsection is given in table 2.3, illustrating that none of the SE materials fulfills all required properties. While in laboratory research, material synthesis typically takes place in house, not many suppliers actually offer commercially available SE materials. Among the most prominent examples are Ampcera<sup>TM</sup>,<sup>19</sup> the NEI Corporation<sup>20</sup>, and the Ohara Corporation<sup>21</sup>. Further issues come up when combined with electrode materials, as will be explained in the following subsection.

#### 2.2.4 Interfaces

The materials and interfaces in an ASSB play a significant role for proper functionality of the ASSB. Degradation induced by electrochemical instability or loss of mechanical contact have been among the main impediments for the successful implementation of large-format ASSBs based on inorganic SEs so far (KERMAN ET AL. 2017). As illustrated by KERMAN ET AL. (2017), the key failure modes are impedance growth and electrical shorting: Contributing factors for impedance growth are loss of interfacial contact at the electrode/current collector interface or at the electrode/SE interface (LIU ET AL. 2017), oxidation or reduction of SEs into ionically insulating phases (WENZEL ET AL. 2016; ZHU

<sup>19</sup> <http://ampcera.com/> (Visited on December 10, 2019)

<sup>20</sup> <https://www.neicorporation.com/products/batteries/solid-state-electrolyte/> (Visited on December 10, 2019)

<sup>21</sup> <https://www.oharacorp.com/lic-gc.html> (Visited on December 10, 2019)

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Table 2.3: Overview of the discussed solid electrolytes with typical total ionic conductivity  $\sigma_i$ . Adapted from ZHANG ET AL. (2018) and FAN ET AL. (2018).

Material	Structure	$\sigma_i$ in S/cm	Advantages	Disadvantages	
Polymer	PEO polymer chains	$<10^{-5}$	stable against Li	ionic conductivity, oxidation stability	
Oxide	LIPON	amorphous	$<10^{-5}$	stable against Li	ionic conductivity
	LAGP	glass-ceramic	$5 \times 10^{-3}$		expensive (Ge)
	LATP	glass-ceramic	$1.3 \times 10^{-3}$	stable in air	unstable against Li
	LLTO	crystalline	$<10^{-4}$		unstable against Li, grain boundary resistance
	LLZ	crystalline	up to $10^{-3}$	stable against Li	sintering temperature, sensitive to humidity
Sulfide	LPS	glass-ceramic	$1.7 \times 10^{-2}$	ionic conductivity	electrochemical stability, reactivity with humidity
	LPSI	glass-ceramic	$1 \times 10^{-3}$	—"—	—"—
	LPSCI	crystalline	$1.3 \times 10^{-3}$	—"—	—"—
	LSiPSCI	crystalline	$2.5 \times 10^{-2}$	—"—	—"—
	LGPS	crystalline	$1.2 \times 10^{-2}$	—"—	—"—, expensive (Ge)

ET AL. 2016)<sup>22</sup>, or the loss of Li-ions into side reactions (VARZI ET AL. 2016). Electrical shorts can occur due to fracture of the SES during operation (KERMAN ET AL. 2017), dendritic growth through defects (NAGAO ET AL. 2013; REN ET AL. 2015a), and oxidation or reduction of SE into electrically conducting phases (HARTMANN ET AL. 2013). Furthermore, poor mechanical assembly or migration of electrode depositions around the separator edges can lead to electrical short circuits (TANAKA 2016). These challenges have to be taken into account when considering manufacturing approaches, which will be the focus of the next section.

<sup>22</sup> Also the electrode materials can change into ion or electron isolating phases (KERMAN ET AL. 2017).

### 2.3 Manufacturing Approaches and Scale-Up

So far, the above described issues of ASSBs with inorganic SEs have hindered successful production of large-format cells with performance competitive to conventional LIB. Based on the previous considerations, implications for ASSB battery design will be deduced in subsection 2.3.1. The subsequent subsections will give an overview on attempts which have been published on fabricating SES (subsection 2.3.2), composite cathode (subsection 2.3.3), and anode layers (subsection 2.3.4), or even battery cells (subsection 2.3.5) using oxide- or sulfide-based SE. Finally, approaches for ASSB scale-up will be summarized in subsection 2.3.6.

#### 2.3.1 All-Solid-State Battery Design

Considerations on the required thicknesses and composition of the ASSB components to be competitive with conventional LIBs can be found in WU ET AL. (2016): When employing contemporary anode materials (e.g. graphite, cf. subsection 2.2.2), the ASSB cannot exceed the specific energy and energy density of a conventional LIB galvanic cell (JANEK & ZEIER 2016). In contrast, when employing a lithium metal anode (cf. subsection 2.2.2), the SES break-even thickness<sup>23</sup> was found to be directly proportional to the active material loading in the cathode. For instance, an ASSB with a typical cathode thickness of 60  $\mu\text{m}$  and a NCA (cf. subsection 2.2.3) active material loading of  $M_c = 19.5 \text{ mg/cm}^2$  would require a SES layer with a thickness below 125  $\mu\text{m}$  to exceed the energy density of a comparable LIC with graphite anode<sup>24</sup>. The break-even thickness for the specific energy depends on the density of the SE material used and may be significantly lower (e.g. below 45  $\mu\text{m}$  for a garnet-type SE, cf. subsection 2.2.3). From a power perspective (equation 2.9), the maximal thickness of the SES layer is calculated to be  $d_s = 16.7 \text{ }\mu\text{m}$  for a SE total ionic conductivity of  $\sigma_i = 10^{-4} \text{ S/cm}$  under typical LIB operating conditions (i.e.  $J = 3 \text{ mA/cm}^2$ ) if the voltage drop across the SES must not exceed  $\Delta V = 50 \text{ mV}$ . For higher ionic conductivities, thicker SES layers can be tolerated—depending on the

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<sup>23</sup> The *break-even thickness* in this context is the critical value for an ASSB with lithium metal anode to have an improved energy density or specific energy in comparison to a conventional LIB, cf. equation 2.4 and equation 2.5.

<sup>24</sup> For details on the underlying assumptions, please refer to the cited reference.

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other performance measures (e.g. energy density or safety, cf. subsection 2.1.2). These considerations on the ASSB cell design—in particular the required layer thicknesses—need to be taken into account when analyzing manufacturing approaches for ASSB components.

### 2.3.2 Solid Electrolyte Separator Fabrication

In laboratory setups, usually the SE powders are compressed to form pellets, typically resulting in thick ( $>100\ \mu\text{m}$ ) SES membranes on which the active materials are applied in an additional pressing or coating step (KATO ET AL. 2012; HU 2016). However, in order to be competitive with conventional LIC, thinner SES layers are required (MCCLOSKEY 2015; WU ET AL. 2016). KERMAN ET AL. (2017) outline different processing routes<sup>25</sup> to fabricate dense SES layers with thicknesses below  $100\ \mu\text{m}$ , as illustrated in figure 2.3.

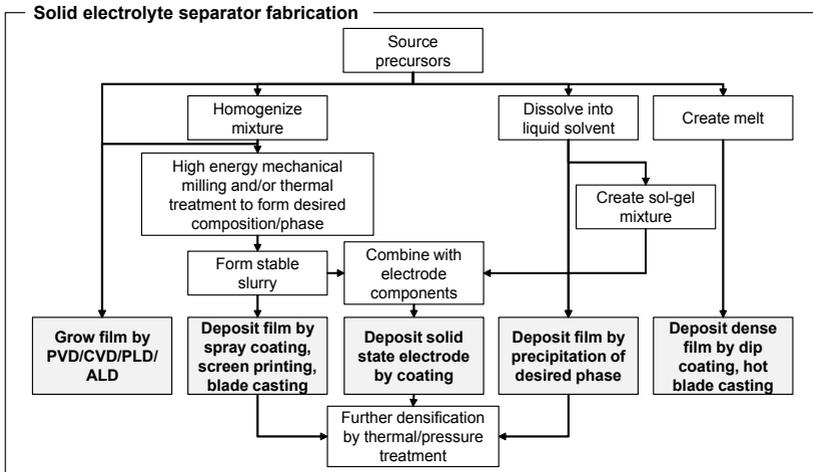


Figure 2.3: Process flow diagram for creating dense solid electrolyte separator layers with thicknesses  $<100\ \mu\text{m}$ . Adapted from KERMAN ET AL. (2017).

<sup>25</sup> Note that the focus of this thesis is not on materials synthesis, i.e. the processes for preparation of SE particles from the precursor materials are only included for the sake of completeness.

The first route (figure 2.3, left branch) describes the deposition of a layer directly from the SE precursors or powders using methods typically applied in thin-film battery fabrication: For LPS, room temperature pulsed laser deposition (PLD) has been successfully applied to fabricate layers in the range of several  $\mu\text{m}$  with a total conductivity of  $2.8 \times 10^{-4} \text{ S/cm}$ . Several attempts have been made to fabricate thin-film LLZ layers, e.g. using atomic layer deposition (ALD) (KAZYAK ET AL. 2017), chemical vapor deposition (CVD) (LOHO ET AL. 2017), PLD (KIM ET AL. 2013), or sputter deposition (LOBE ET AL. 2016). However, none of these approaches was successful in fabricating layers with ionic conductivities exceeding  $10^{-5} \text{ S/cm}$ . Most of these technologies are very sensitive to the substrate morphology and temperature (KERMAN ET AL. 2017), which is in particular critical when depositing films directly onto one of the electrodes. Recently, the aerosol deposition method (ADM) has been applied to create LLZ films, with ionic conductivities approaching  $10^{-4} \text{ S/cm}$  (HANFT ET AL. 2017). BAEK ET AL. (2014) also discuss further spraying techniques, such as plasma spray or high velocity oxygen fuel spraying (HVOF), although the authors state that the high processing temperatures probably lead to structural decomposition of the components. Since most of the addressed technologies do not require a subsequent densification step, the following hypothesis is made:

**Hypothesis H 3: Vapor deposition or spray coating techniques are promising technologies for SES fabrication.**

An increasing number of publications have been investigating the solvent-based fabrication of SES layers (figure 2.3, middle branches): Here, the SE precursors or particles are dissolved into a liquid, e.g. to form a slurry or sol-gel mixture which is subsequently used to cast or coat the SES layer<sup>26</sup>. Wet coating or tape casting<sup>27</sup> have been widely established to fabricate layers<sup>28</sup> with thicknesses below  $100 \mu\text{m}$ . While sol-gel dip or spin coating were successfully applied for

<sup>26</sup> Similarly, a composite electrode could be fabricated, as discussed in the following subsections.

<sup>27</sup> Here, the terms *wet coating* and *tape casting* are used to summarize a variety of similar coating or casting technologies typically differentiated by the application tool (e.g. comma bar, slot die, or rotating roller) and the substrate, i.e. another cell component (coating) or a carrier tape (casting).

<sup>28</sup> Layers with sulfide-based SEs have been manufactured, for instance, by INADA ET AL. (2009), NAM ET AL. (2015), OH ET AL. (2017), SAKUDA ET AL. (2017), YAMAMOTO ET AL. (2018), NAM ET AL. (2018), and ATES ET AL. (2019); layers with oxide-based SEs by YI ET AL. (2016), YI ET AL. (2017), FU ET AL. (2017b), XU ET AL. (2018b), and HITZ ET AL. (2019).

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sulfides (WANG ET AL. 2013; WANG ET AL. 2016), only poor ionic conductivities could be achieved for LLZ ((BITZER ET AL. 2016), (TADANAGA ET AL. 2015), (CHEN ET AL. 2014)). Sulfide-based layers with varying thicknesses have also been fabricated using electrophoretic deposition (EPD) (AZUMA ET AL. 2017). Further suggested technologies include spray coating and screen printing (BAEK ET AL. 2014; KERMAN ET AL. 2017). Extrusion or co-extrusion (with one or two electrodes) have also been suggested to reduce the solvent content and achieve lower porosities (JOHNSON & JOHNSON 2014; BAE ET AL. 2016).

Since the solvent evaporation typically results in a porous layer, further densification is required by compression or heat treatment. While the ductile nature of sulfide-based SEs allows for densification by simple pressure application (SAKUDA ET AL. 2013), oxide-based SEs usually require a subsequent temperature treatment (i.e. sintering, cf. subsection 2.2.3). In this case, particular attention must be paid to the stability of all components to be sintered (e.g. co-sintering with cathode active materials, cf. next subsection). Sintering additives (such as  $\text{Li}_3\text{BO}_3$  (LBO) or  $\text{Al}_2\text{O}_3$ ) have been used to reduce the sintering temperatures down to  $790^\circ\text{C}$  (OHTA ET AL. 2014). Further approaches include alternative sintering methods, such as spark plasma sintering (SPS) or field assisted sintering which could particularly reduce the required sintering times (BAEK ET AL. 2014; ZHANG ET AL. 2014).

The last suggested route (figure 2.3, right branch) is the preparation of a SES layer directly from a melt, which corresponds to an established process in the glass industry (KERMAN ET AL. 2017).

A selection of relevant SES fabrication approaches for different oxide- or sulfide-based SE materials is summarized in table 2.4<sup>29</sup>. According to the considerations above, only technologies were included where the final layer (after post-processing) had a thickness below  $100\ \mu\text{m}$  and a total ionic conductivity above, or at least close to,  $10^{-4}\ \text{S/cm}$ . Where possible, also the substrate, as well as subsequent densification steps were supplemented<sup>30</sup>.

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<sup>29</sup> Manufacturing approaches for hybrid SEs can be found in KELLER ET AL. (2018).

<sup>30</sup> Note that many of these attempts are rather proof-of-concepts that did not combine the resulting layers with electrodes to fabricate actual ASSBs.

## 2.3 Manufacturing Approaches and Scale-Up

Table 2.4: Overview of the discussed fabrication approaches for inorganic solid electrolyte separator layers with thickness  $d_s$  below 100  $\mu\text{m}$  (after densification) and resulting total conductivities in the order of  $10^{-4}$  S/cm or higher.

Technology	Material	Substrate	Densification	$d_s$ in $\mu\text{m}$	Ref.
Tape casting	LGPS	Al foil/ plastic mesh	100 MPa, 100 °C	100	INADA ET AL. (2009)
	LPS/ LGPS	Ni foil	pressing	70	NAM ET AL. (2015), OH ET AL. (2017)
	LPS	Cu foil	330 MPa to 410 MPa	70	SAKUDA ET AL. (2017)
	LPS	free-standing / electrode	110 MPa, 225 °C	59	YAMAMOTO ET AL. (2018)
	LPSCI	graphite anode	370 MPa (cell pressing)	30	NAM ET AL. (2018)
	LPS	NMC cathode/ Mylar foil	pressing	20	ATES ET AL. (2019)
	LLZ	Mylar foil	>1100 °C (after lamination onto porous support)	14 to 35	FU ET AL. (2017b), XU ET AL. (2018b), HITZ ET AL. (2019)
	LLZ	Mylar foil	>1080 °C	25 to 28	YI ET AL. (2016), YI ET AL. (2017)
Dip coating	LPS	Ni foil	200 °C	8 to 50	WANG ET AL. (2016)
Spin coating	LGPS	glass	240 °C	0.13	WANG ET AL. (2013)
EPD	LPS	glass	220 °C, 330 MPa	10 to 100	AZUMA ET AL. (2017)
PLD	LPS	silicon/ glass	200 °C	3	SAKUDA ET AL. (2010)
ADM	LLZ	gold/ silicon	200 °C to 700 °C	5	HANFT ET AL. (2017)

### 2.3.3 Composite Cathode and Half-Cell Fabrication

Since the energy density and specific energy are mainly governed by the amount of cathode active material in the battery cell, the thickness and composition of the cathode needs to be carefully tuned. While thin-film deposition of cathodes onto SES pellets, e.g. by PLD, has been employed to fabricate ASSB (OHTA ET AL. 2012), the resulting active material loading is several orders of magnitude too low for large-format applications. Different approaches for preparing thicker composite cathodes with high active material loading are illustrated in figure 2.4. Since the interface with the SES layer is of major importance, the fabrication of half-cells (composite cathode and SES) will also be discussed.

The first concept (figure 2.4, left image) describes the coating of a composite cathode mixture onto a current collector or fabrication of a free-standing layer and subsequent joining with the current collector, similar to conventional LIC production. Intimate contact between particles can be improved by homogeneous mixing of cathode and SE particles or the reduction of particle sizes (NAM ET AL. 2018; WANG ET AL. 2019). Next to dry powder pressing, slurry based processes have been most widely established. Here, cathode powders are dissolved with SE precursors or SE powders (ITO ET AL. 2014; OH ET AL. 2017), as well as binders and conductive additives and a solvent. Core-shell coatings of cathode active materials (such as NCA and NMC), e.g. with LTO (SEINO ET AL. 2011),  $ZrO_2$  (MACHIDA ET AL. 2012),  $Li_2O-ZrO_2$  (ITO ET AL. 2014), or  $LiNbO_3$  (SAKUDA ET AL. 2017), are often applied to improve the interface properties with the SE. After mixing, the layer is cast onto a carrier tape or coated onto a current collector<sup>31</sup>. A different fabrication concept was suggested by KIM ET AL. (2017), as illustrated in figure 2.4 (middle image): Here, a porous cathode, prepared by conventional LIC electrode fabrication processes, was dip coated and infiltrated by a sulfide-based SE slurry. Next to solvent-based methods, also the ADM has been used, for instance for co-deposition of LNMO cathode and LATP SE particles onto a current collector (IRIYAMA ET AL. 2018).

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<sup>31</sup> This approach has been used, for instance, by ITO ET AL. (2014), YAMAMOTO ET AL. (2017), OH ET AL. (2017), SAKUDA ET AL. (2017), LEE ET AL. (2017), CHOI ET AL. (2017), NAM ET AL. (2018), YAMAMOTO ET AL. (2018), CHOI ET AL. (2018), ATE ET AL. (2019).

## 2.3 Manufacturing Approaches and Scale-Up

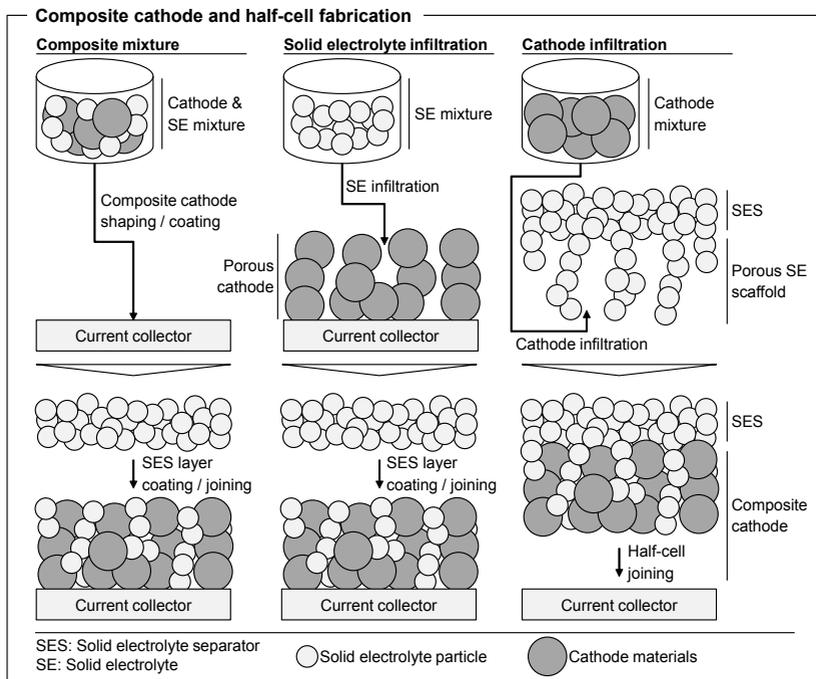


Figure 2.4: Schematic illustration of different approaches for composite cathode and half-cell fabrication. Left: Direct shaping or coating of a layer based on a mixture of cathode and SE materials, e.g. ITO ET AL. (2014), OH ET AL. (2017). Middle: Infiltration of a SE mixture into a porous cathode, e.g. KIM ET AL. (2017). Right: Infiltration of a cathode mixture into a porous SE scaffold, e.g. WACHSMAN ET AL. (2014), REN ET AL. (2017).

After fabrication of the composite cathode layer, a half-cell can be fabricated by joining or coating with the SES layer (ATES ET AL. 2019).

For most sulfide-based SEs, the layer fabrication process is typically followed by a cold or hot pressing step to reduce the porosity and obtain more intimate contact between the particles. Pre-compression of the cathode layer before joining with the SES can improve performance (YAMAMOTO ET AL. 2017). However, despite the ductility of the sulfide-based SE, achieving close-to-zero porosity has turned out to be challenging (SAKUDA 2018), and pressures too high easily lead to pulverization or cracking of the particles (YAMAMOTO ET AL.

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2017). Pre-coating of cathode active materials with SE materials or smaller particle sizes have been suggested as a potential solution (SAKUDA ET AL. 2011; SAKUDA ET AL. 2016).

Obtaining intimate contact between cathode particles and oxide-based SEs seems an even bigger challenge: Typically, temperatures in the order of 1000 °C are required to reduce the grain boundary resistance between the SE particles (cf. subsection 2.2.3). However, most cathode active materials have shown to form undesired side products with the SE materials when co-sintered at these elevated temperatures: Decomposition of both LATP and LLZ with different high voltage cathodes (such as LNMO) has been observed at sintering temperatures above 600 °C (MIARA ET AL. 2016). In contrast, LATP and NMC could be sintered up to a temperature of 700 °C, although mutual diffusion remains an issue (KATO ET AL. 2016a). By introducing sintering aids (e.g. LBO), LLZ and NMC could be co-sintered at a temperature of 700 °C (WANG ET AL. 2019). Due to the challenge to co-sinter oxide-based SES layers with composite cathodes, most fabrication approaches for oxide-based ASSB have aimed at preparing dense SES layers with a high temperature sintering step and subsequent joining or coating with a cathode composite. For instance, screen-printing has been applied to coat the cathode onto a sintered LLZ pellet, followed by subsequent low temperature co-sintering in order to reduce the interface resistance of the two layers (OHTA ET AL. 2013; FENG ET AL. 2017). In a similar fashion, the ADM has been used to fabricate a composite cathode (LATP with NMC) on LLZ (KATO ET AL. 2016b). While these approaches seem plausible for deposition onto thick (>100 µm) pellets, handling of large-format free-standing SES layers has turned out to be challenging because of the brittleness of the sintered layers (HITZ ET AL. 2019). This makes the subsequent joining or coating with the composite cathode a highly critical endeavor (KERMAN ET AL. 2017).

Therefore, recent progress has been made for a different concept (figure 2.4, bottom left image), adapted originally from solid oxide fuel cell (SOFC) fabrication (MCINTOSH & GORTE 2004): Here, a porous LLZ catholyte<sup>32</sup> scaffold is fabricated as a supporting layer, for example by tape casting of a green sheet using a LLZ slurry with pore formers (FU ET AL. 2017b; HITZ ET AL. 2019).

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<sup>32</sup> Here, the term *catholyte* is used to describe the SE ion percolating network in the composite cathode.

After solvent evaporation, this layer is coated or laminated with the SES green sheet (without pore formers). During the subsequent sintering step, the pore formers are removed, resulting in a bilayer structure<sup>33</sup> with a thick, porous support ( $\approx 70 \mu\text{m}$ , 60 % porosity) and a thin, dense SES ( $14 \mu\text{m}$  to  $35 \mu\text{m}$ ,  $< 1 \%$  porosity) (FU ET AL. 2017b; HITZ ET AL. 2019). The three-dimensional sintered host structures<sup>34</sup> enable the subsequent infiltration of the cathode materials, for example by multiple subsequent sol-gel coating and calcination steps<sup>35</sup> (SHOJI ET AL. 2016; REN ET AL. 2017). Sintering aids have been used to reduce the interface resistance of the cathode materials with the sintered LLZ scaffold (SHOJI ET AL. 2016). Finally, the resulting half-cell can be joined with the current collector (and anode).

**Hypothesis H 4: For oxide-based ASSBs, infiltration of cathode materials into a porous, sintered SE host is a promising approach.**

A summary of the discussed half-cell fabrication approaches with corresponding literature examples is given in figure 2.5.

### 2.3.4 Anode Fabrication

As outlined in subsection 2.2.2, the focus of this subsection will be on the fabrication of a lithium metal anode<sup>36</sup>. The integration of a lithium metal anode has been challenging, especially due to the interface at the SES layer. An anode-free fabrication has been successfully implemented for thin-film batteries, where the lithium anode is generated by plating from the cathode during the first charge (NEUDECKER ET AL. 2000). However, irreversible losses during cycling will easily lead to a capacity fade (QIAN ET AL. 2016). Hence, in most laboratory approaches, typically thick ( $> 100 \mu\text{m}$ ) lithium foils are employed. Lithium foil is usually fabricated by extrusion with subsequent rolling steps to thin down

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<sup>33</sup> Trilayer structures were also fabricated to provide another porous layer on the anode side (*anolyte*) WACHSMAN ET AL. (2014), XU ET AL. (2018b), HITZ ET AL. (2019), cf. next subsection.

<sup>34</sup> Different approaches to obtain a dense SES combined with a 3D LLZ host structure are the preparation of a hole-array pellet using a die with patterned structure (SHOJI ET AL. 2016), or 3D-printing (MCOWEN ET AL. 2018).

<sup>35</sup> Melt-infiltration is often used for sulfur-based cathodes, e.g. FU ET AL. (2017b), XU ET AL. (2018b), HITZ ET AL. (2019). These are, however, not in the scope of this thesis (cf. section 2.2).

<sup>36</sup> The considerations on manufacturing of composite cathodes in the previous subsection could also be applied to composite anodes, e.g. with graphite as active material.

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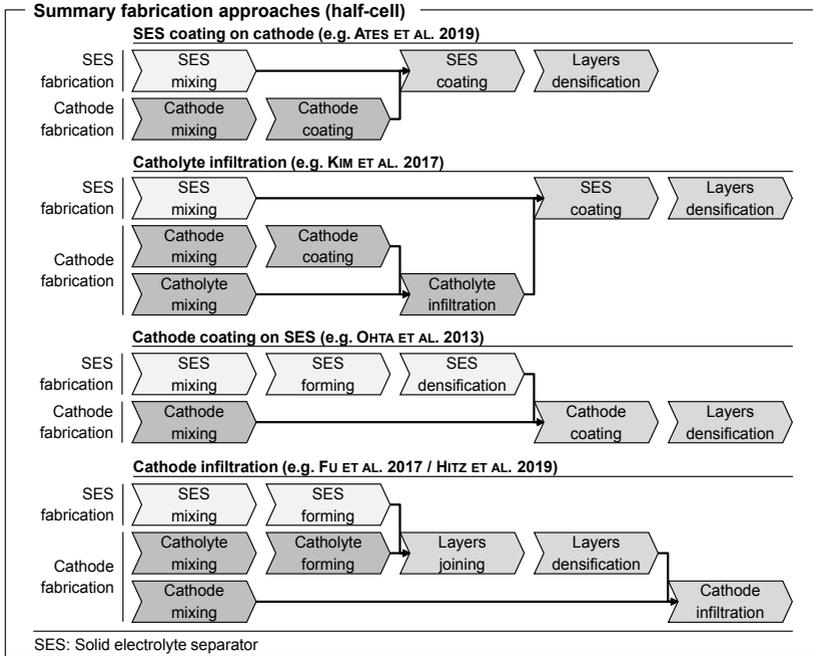


Figure 2.5: Summary of the discussed approaches for half-cell fabrication.

the foil to the desired thickness (HOVSEPIAN 1973; BOUCHARD ET AL. 1996). However, this process is challenging for thinner layers due to the adhesiveness and reactivity of lithium. Hence, so far, only few suppliers offer rolled lithium foils with thicknesses approaching 20  $\mu\text{m}$ , which is a requirement for competitive energy density (JANEK & ZEIER 2016). Melt coating, for instance onto a Cu current collector, seems to be another option to fabricate thin lithium layers (KOKSBANG & JENSEN 1992; BELANGER ET AL. 1993), although the reactivity of molten lithium calls for elaborate safety precautions (MEYER 1957). Similar conclusions can be drawn for vacuum evaporation (BRODD 1996; BASAPPA ET AL. 2017a). Electrochemical deposition has also been applied to plate lithium onto Cu substrates (SWONGER 2015; MASHTALIR ET AL. 2018), although layer growth is highly sensitive to the substrate properties (KIM ET AL. 2019).

**Hypothesis H 5: Manufacturing and processing of thin lithium metal films are critical both technically and economically.**

A homogeneous interface and intimate contact are key for cycling at high current densities (HITZ ET AL. 2019). Interface resistance at the anode side can be reduced by ALD coating with  $\text{Al}_2\text{O}_3$  or ZnO (HAN ET AL. 2016; XU ET AL. 2018b), or by surface polishing or direct vacuum evaporation onto the SES surface (BASAPPA ET AL. 2017a). Lithium metal alloys (e.g. formed by electron beam evaporation of 20 nm Al) or polymer interlayers (e.g. by coating of 2  $\mu\text{m}$  PEO) have been integrated to homogenize the interface of metallic lithium with LLZ (FU ET AL. 2017b; FU ET AL. 2017a) or LPS (XU ET AL. 2018a; ATES ET AL. 2019). Similar to the fabrication of a porous catholyte scaffold (cf. previous subsection), a porous LLZ matrix could also be employed at the anode side to enhance the effective surface area and accommodate the volume changes during cycling. Melt infusion has been used to infiltrate metallic lithium into these porous LLZ layers, typically also in combination with appropriate surface treatment (YANG ET AL. 2018; XU ET AL. 2018b).

### 2.3.5 Cell Assembly

In contrast to most laboratory approaches, where pellet or coin cells are used, not many attempts have been published on the assembly of large-format ASSBs. However, the number of patent applications on ASSB cell assembly and packaging has been rapidly increasing during the last years<sup>37</sup>.

Similar to conventional LIC production (cf. figure 1.2), punching (SATOU 2017) or laser cutting (JOHNSON & JOHNSON 2014) have been suggested for cutting of ASSB components. While for mechanical cutting, the adhesiveness and reactivity of metallic lithium can pose an issue, contaminations on the lithium surface can lead to spontaneous ignition during laser cutting, even in dry atmosphere (JANSEN ET AL. 2018). It is noteworthy that not only cutting of single electrode and SES sheets was suggested, but also cutting of half cells (SES layer on electrode) or galvanic cells (anode, SES, cathode). This requires

<sup>37</sup> Examples are HASEGAWA ET AL. (2018a), HASEGAWA ET AL. (2018b), HASEGAWA & KUROMIYA (2018), KIM ET AL. (2018), YOON ET AL. (2018), OKAMOTO ET AL. (2019).

elaborate design of the sheets to be cut, e.g. a patterned coating to hinder short circuiting along the cut line (JOHNSON & JOHNSON 2014).

Stacking of multiple galvanic cells has mainly been carried through manually on laboratory scale, for both parallel (e.g. ITO ET AL. (2014)) and bipolar stacking (e.g. NAM ET AL. (2015)). Due to the brittle nature of SE and composite cathode layers with low porosity (ATES ET AL. 2019), conventional winding or folding processes seem rather unlikely. Nonetheless, addition of polymeric binders allows for a certain flexibility (RIPHAUS ET AL. 2018). Pressing or heat treatment of the assembled cell stack can be beneficial to enhance contact between the layers or to remove undesired binder residuals (YAMAMOTO ET AL. 2018).

Up to date, not many publications have been dealing with packaging of SSBs. Most laboratory cells are compressed in a special setup to ensure sufficient pressure application during cycling (KATO ET AL. 2012; HU 2016). For ASSB with larger lateral dimensions, a pouch bag casing is usually employed (e.g. by ITO ET AL. (2014) or NAM ET AL. (2018)). Large-format prototype cells have been presented, for instance, by Toyota Motor Corporation<sup>38</sup> and Samsung<sup>39</sup>. An overview of different prototype cells demonstrated by industrial companies was given by SUN ET AL. (2017). However, only few publications have actually described the role of variable or fixed pressure during cycling of ASSBs (ZHANG ET AL. 2017; KOERVER ET AL. 2018).

### 2.3.6 Considerations for Scale-Up

For polymer based and thin-film SSBs, production on pilot or industrial scale has been successfully implemented by wet coating (polymers) or sputtering (thin-film) electrode and SES layers on top of each other (PATIL ET AL. 2008; GRAPE 2015). However, for large-format ASSB with inorganic SEs, only few attempts have been reported on up-scaling of production. MA ET AL. (2016) developed a method for large-scale production of LATP materials. XU ET AL. (2018b) and HITZ ET AL. (2019) produced several meters of LLZ layers by roll-to-roll tape casting on a pilot facility, although further processing took place

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<sup>38</sup> [https://tech.nikkeibp.co.jp/dm/english/NEWS\\_EN/20101122/187553/](https://tech.nikkeibp.co.jp/dm/english/NEWS_EN/20101122/187553/) (Visited on December 10, 2019)

<sup>39</sup> <https://www.sait.samsung.co.kr/saithome/research/battery.do> (Visited on December 10, 2019)

on laboratory scale. Several start-up companies have claimed fabrication of ASSBs on pilot scale; prominent examples are, for instance, Solid Power<sup>40</sup> and QuantumScape<sup>41</sup>.

Theoretical considerations on up-scaling were presented by various authors. BAEK ET AL. (2014), XIN ET AL. (2017), and HAO ET AL. (2018) discuss different production technologies for ASSB layer fabrication and densification: While slurry-based processes (such as tape casting) typically require pressure or temperature treatment for compaction, thin-film processing (such as sputtering, CVD, PLD, or ALD) or spray coating technologies (such as plasma spraying, HVOF, cold spray, or ADM) could potentially create layers without the need for further densification. Thin-film processing, however, raises concerns with regard to scalability. Spray coating is also suggested by HU (2016), who further illustrates a slurry-based production route where SES and electrodes are directly coated on top of each other (figure 2.6, upper image). Similar, yet more elaborate process layouts were described in a patent by JOHNSON & JOHNSON (2014), where multiple coating and calendaring steps are suggested for ASSB galvanic cell fabrication. For instance, coating of a composite cathode on a current collector or substrate is succeeded by coating the SES and, finally, lamination or evaporation of anode and/or anode current collector. Similarly, the SES can be coated onto the anode, followed by coating of the composite cathode and lamination of the cathode current collector. The same patent also describes the co-extrusion of cathode and SES and subsequent lamination or evaporation of the anode and current collectors (figure 2.6, bottom image). As discussed in the previous subsection, an intermittent anode coating can facilitate the subsequent layer cutting process. Co-extrusion of all three layers (cathode, SES, and anode) and lamination of the current collectors was also suggested.

Targets, challenges and potential solutions for production of lithium anodes and various types of SES layers (polymer, hybrid, inorganic) are roughly outlined in a roadmap by THIELMANN (2017): For inorganic SESs, next to issues with interfaces and cell design, sintering temperatures (for oxides) and processing atmospheres (in particular for sulfides) raise concerns, as also postulated within

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<sup>40</sup> <https://solidpowerbattery.com/> (Visited on December 10, 2019)

<sup>41</sup> <https://www.quantumscape.com/> (Visited on December 10, 2019)

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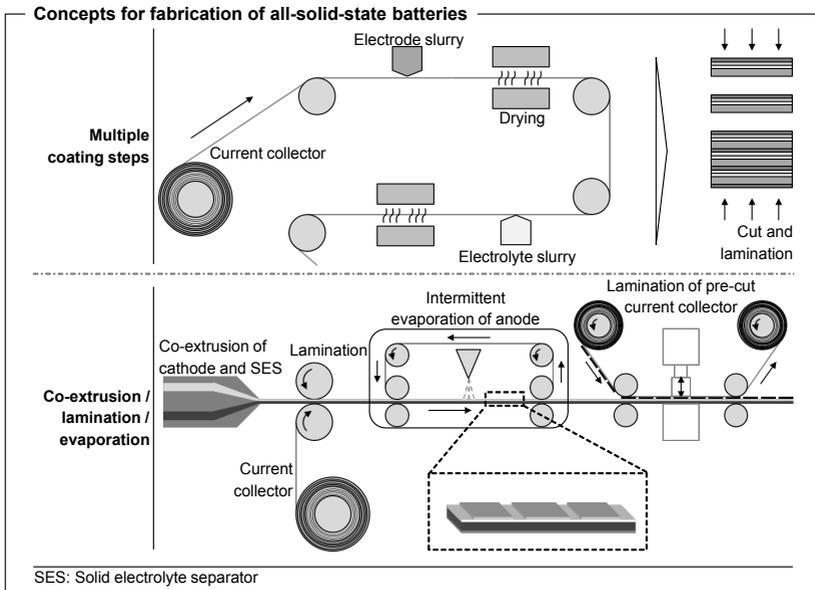


Figure 2.6: Exemplary concepts for large-scale fabrication of ASSBs. Adapted from HU (2016) and JOHNSON & JOHNSON (2014).

the hypotheses H 1 and H 2. Further bottlenecks are the large-scale deposition of SES layers, as well as subsequent handling and stacking. Within the frame of a life cycle assessment, TROY ET AL. (2016) outline possible production steps for the fabrication of oxide-based ASSB pouch cells. Here, two subsequent tape casting and sintering steps are suggested for SES layer and cathode composite, respectively, followed by a welding process to join the resulting half cell with lithium foil and current collectors (figure 2.7). Based on these assumptions, conclusions are drawn on the energy consumption and the criticality of different raw materials, such as lithium, lanthanum, and zirconium. Estimations on the material cost were presented by MCCLOSKEY (2015) and SCHMUCH ET AL. (2018), indicating high uncertainties associated in particular with SE and lithium material cost. With regards to the up-scaling towards pilot and industrial production, these considerations lead to the following hypothesis:

**Hypothesis H 6: Material supply and cost are critical for ASSB production.**

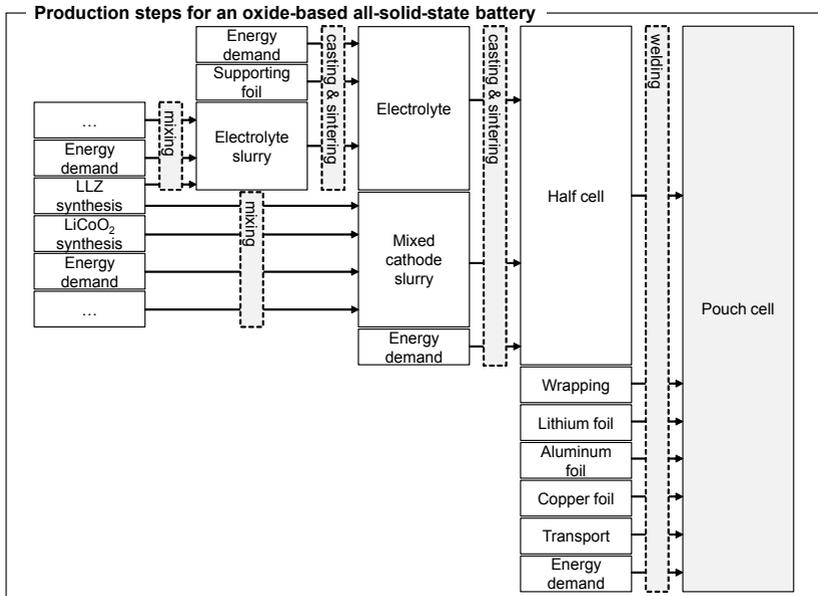


Figure 2.7: Exemplary production steps with input and output for an oxide-based ASSB pouch cell. Adapted from TROY ET AL. (2016).

## 2.4 Conclusion

The results of this chapter can be summarized as follows: Based on the fundamental properties and requirements on ASSBs, the overview of materials and interfacial challenges served as a basis in order to categorize the different fabrication approaches for ASSB components and cells. To conclude, no large-format ASSB competitive to conventional LIBs and producible on an industrial scale has been reported yet. In order to facilitate strategic planning for the industrial fabrication of ASSBs and to derive answers to the hypotheses defined within this chapter, the following challenges need to be addressed (figure 2.8):

- The upscaling of ASSB production is subject to a high uncertainty, caused by a multitude of different SE materials and an ambiguous cell design.
- Furthermore, the current literature lacks an overview on possible production technologies and their respective challenges. While a large variety

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and diversity of production technologies for fabrication of ASSB components has been applied on the laboratory scale, the implementation of several of these technologies in an industrial environment is questionable.

- Associated with the different material combinations and production technologies are the challenges at the interfaces between the particles and components of an ASSB. In particular, these interactions occur differently for varying production technologies.
- Current knowledge on ASSBs comes mainly from laboratory experiments, especially in solid-state chemistry and physics. Insufficient knowledge from a production engineering perspective has hindered Simultaneous Engineering for ASSB development so far.
- Finally, in order to successfully launch the ASSB as a product on the market, performance and cost must be competitive to conventional LIBs. However, due to ongoing development efforts, the current performance and cost of LIB will soon be outdated (SCHMUCH ET AL. 2018). Hence, a profound comparison should also take into account future advances in conventional LIB technology (“running target”).

The resulting complexity needs to be tackled by a systematic and structured procedure (CAMPBELL 1988). Hence, in the following, the state of the art on strategic planning of technology chains will be analyzed with regards to the requirements on a methodological framework derived from the underlying research problem.

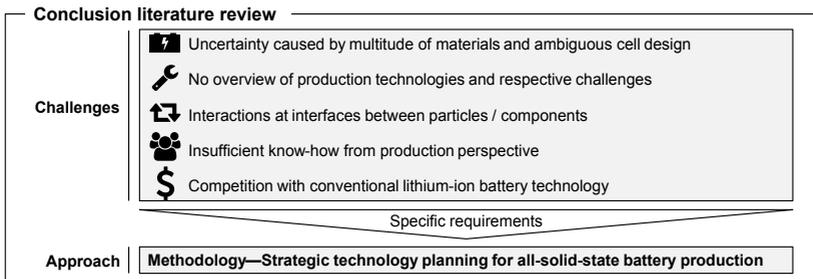


Figure 2.8: Challenges resulting from the current state of research on ASSB scale-up.

### 3 Strategic Planning of Technology Chains

Following the considerations on all-solid-state batteries (ASSBs) within the previous chapter, an overview of methods applied in strategic planning of technology chains will be given here. For this purpose, requirements on a methodological procedure will be deduced in section 3.1, followed by a brief introduction to technology management in section 3.2. This serves as a basis to review and assess the state of the art on strategic planning of technology chains in section 3.3. The resulting action required will be derived in section 3.4.

#### 3.1 Methodology Requirements

In this section, requirements on the methodological procedure for strategic planning of technology chains for ASSB production will be derived, as depicted in figure 3.1: The specific requirements which are based on the underlying research problem will be explained in subsection 3.1.1. Furthermore, a number of formal requirements need to be fulfilled, which will be explicated in subsection 3.1.2.

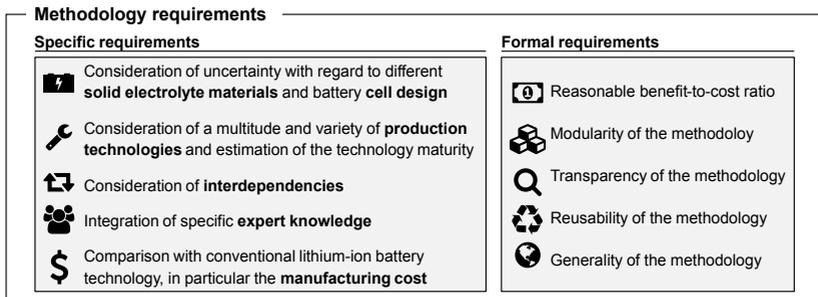


Figure 3.1: Specific and formal requirements on the methodology for strategic planning of technology chains for ASSB production.

##### 3.1.1 Specific Requirements

From the considerations on the current status of ASSBs and the resulting challenges summarized in section 2.4, the following specific requirements on the methodology can be derived:

### 3 Strategic Planning of Technology Chains

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- *Consideration of uncertainty with regards to different SE materials and battery cell design:* As summarized in section 2.2, a large variety of different SE materials is currently discussed in scientific literature. Until today it is not clear which materials are best suited to match the performance measures explained in subsection 2.1.2. Furthermore, the specific product and component design is strongly dependent on the materials combination (cf. section 2.3) and, thus, remains ambiguous up to date. To enable a Simultaneous Engineering approach (cf. section 1.3), the application of Design for Manufacture and Assembly (DFMA)<sup>1</sup> concepts needs to be promoted to adjust the cell design under consideration of the capabilities of different production technologies.
- *Consideration of a multitude and variety of production technologies and estimation of the technology maturity:* On the laboratory scale, a multitude of different production technologies has been applied to process ASSB components (cf. section 2.3). Therefore, the methodology must be capable to comprehensively analyze and compare a large amount of technologies, in particular considering the respective technology maturity and scalability. Additionally, a large variety of engineering disciplines is involved in battery production (WESTERMEIER ET AL. 2014), covering aspects of chemical engineering, process engineering, manufacturing and assembly technology, and electrical engineering. This multiplexity of production technologies and disciplines must be covered by the methodology.
- *Consideration of interdependencies:* Different types of interdependencies need to be considered during planning of technology chains: This entails the interdependencies between the different materials and components, as well as interactions with and between different production technologies.
- *Integration of specific expert knowledge:* Since most of the knowledge on ASSBs so far has been derived from theoretical and experimental considerations on the laboratory scale, only few people have a general expertise on all subject areas of this thesis (cf. section 1.3). Therefore, knowledge

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<sup>1</sup> *Design for Manufacture and Assembly* “[...] provides a systematic procedure for analysing proposed designs from the point of view of assembly and manufacture [resulting] in simpler and more reliable products which are less expensive to assemble and manufacture” (BOOTHROYD 1994).

from a large number of experts with specific know-how on solid state chemistry and physics, production engineering, and strategic technology planning needs to be integrated within an interdisciplinary approach.

- *Comparison with conventional LIB technology, in particular the manufacturing cost:* The identification of critical process steps is among the major requirements (cf. section 1.3). Since the LIB market is a strongly dynamic field (SCHMIDT ET AL. 2017), the final success of the ASSB will be dependent on whether it can be produced at costs competitive to the future price of a LIB. Due to the very specific requirements of ASSB materials (cf. section 2.2), a simple adaption of cost structures from similar systems (e.g. LIB production, ceramics manufacturing) will hardly lead to satisfactory results. Therefore, a detailed analysis of the cost structure and manufacturing processes of ASSB production will be required.

#### 3.1.2 Formal Requirements

The central aspects considered within the framework of this thesis are based on current research on ASSB. This is afflicted with high uncertainties and difficult accessibility of information. Hence, in addition to the specific requirements explained in the previous section, a number of formal requirements results from the underlying research problem<sup>2</sup>:

- *Reasonable benefit-to-cost ratio:* In order to allow for an economically feasible application of the methodology, the associated effort and expenses must be adequate with regards to the achievable benefits.
- *Modularity of the methodology:* This can be facilitated by a modular structure where the level of detail for the individual models is adjustable towards the specific use case. Hence, the modules need to be easily understandable and the interfaces between them must be designed to ensure consistency.

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<sup>2</sup> Similar considerations can be found in adjacent literature, for instance HEIMES (2014, 65ff.) and KNOCHE (2018, 143ff.).

### 3 Strategic Planning of Technology Chains

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- *Transparency of the methodology:* A transparent and systematic approach is required to cope with the information diversity and to ensure a reproducible generation of results. This ensures that critical process steps and cost drivers in ASSB production can be identified.
- *Reusability of the methodology:* As explained in section 1.4, the amount of literature on ASSBs has grown strongly during the last years. Hence, the current state of research on which this thesis is based will soon be outdated. In order to ensure a reusability of the methodology, the modules must be easily extendable, for instance to enable updates with regard to emerging technologies not considered so far.
- *Generality of the methodology:* Due to the targeted use case scenario, the methodology must be specifically tailored towards the requirements and challenges for industrial production of large-format ASSBs. A general validity of the methodology for other products is therefore not given. Nonetheless, the methodology should be easily transferable to related materials, cell designs, and production technologies. This holds particularly true for a comparison with conventional LIB production.

In order to develop a suitable methodology capable of meeting the above defined requirements, the fundamentals of strategic technology management will be explained and an overview about methods employed in strategic planning of technology chains will be given in the following sections. This will help to select suitable approaches which are subsequently combined in a specifically adapted methodology.

## 3.2 Fundamentals

In order to assess the state of the art in strategic planning of technology chains with regards to the previously defined requirements, a brief introduction into strategic technology management and technology chain planning will be given in the following.

### 3.2.1 Framework Technology Management

One of the objectives of this thesis is to assist industrial stakeholders to make profound decisions with regards to strategic investments associated with ASSB

production (cf. subsection 1.3.1). The **technology strategy** results from the corporate and business strategy as a part of the production strategy (figure 3.2, upper image) (ZAHN 1988, p. 527; MARTINI 1995, p. 13). Typically, a differentiation is made by the technology performance (technology leadership vs. technology presence) or the technology timing (technology pioneer vs. technology follower)<sup>3</sup> (SCHULTE-GEHRMANN ET AL. 2011, 69ff.). The technology strategy is closely correlated to the main activities of technology management (cf. section 1.2) (SCHUH ET AL. 2011b, 14ff.). Due to the long-term planning horizon of the underlying research problem (cf. section 1.2), the focus of this thesis is on the activities of technology forecasting<sup>4</sup>, technology planning, and technology assessment (figure 3.2, middle image), as will be described in the following paragraphs.

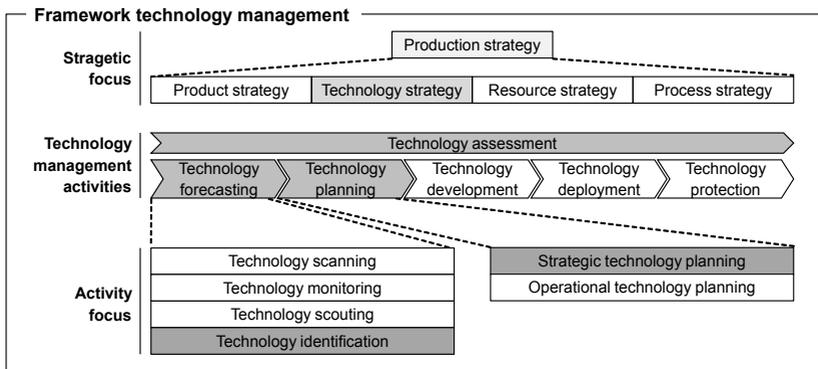


Figure 3.2: Illustration of the interaction between the technology strategy as a part of the production strategy and the basic activities of technology management. Adapted from MARTINI (1995, p. 13), SCHULTE-GEHRMANN ET AL. (2011, p. 86), and GREITEMANN (2016).

A systematic procedure is required to gather information on the multitude and variety of production technologies potentially suitable for ASSB produc-

<sup>3</sup> These strategies are also in close interaction with the market or competitive strategy (i.e. cost leadership, differentiation, or focus) (PORTER 1998, 35ff. SCHULTE-GEHRMANN ET AL. 2011, 78ff.).

<sup>4</sup> *Technology forecasting* can be defined as “the systematic prediction of future performance characteristics and applications of familiar and yet-to-be developed technologies” (TSCHIRKY 1994). The terms *technology intelligence* or *technology foresight* are also often used, cf. GREITEMANN (2016, p. 8) and GIBSON ET AL. (2018) and references therein.

### 3 Strategic Planning of Technology Chains

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tion (cf. section 2.3). **Technology identification** is among the main activities of technology forecasting (figure 3.2, bottom image); the objective of which is to identify developments in relevant technology fields as a basis for strategic decision-making processes (KERR ET AL. 2006; SCHUH ET AL. 2011b, p. 15)<sup>5</sup>. Typical methods include literature and patent analyses, expert interviews, portfolios, the scenario technique, checklists, etc. (LICHTENTHALER 2005, p. 69; WELLENSIEK ET AL. 2011, pp. 134, 151). A methodology specifically designed for the systematic identification of production technologies was developed by GREITEMANN (2016)<sup>6</sup>.

The information determined within technology forecasting is key to the evaluation and selection of suitable technologies during **technology planning** (NAUDA & HALL 1991; SCHUH ET AL. 2011c, p. 192). Relevant input from the technology strategy are, e.g. the desired technological performance, technology sources and the timing of technology application (SCHUH ET AL. 2011c, p. 196). The selection is particularly determined by the information on the products' properties, characteristics, and functionalities (SCHUH ET AL. 2011c, p. 176), which can only vaguely be described within the framework of strategic technology planning (cf. section 1.2) (SCHMITZ 1996, 35f. EVERSHEIM ET AL. 1997). Further input are, for instance, the targeted quantities, markets, and unit costs (SCHUH ET AL. 2011c, p. 176). A methodology for strategic planning of manufacturing technologies was developed by SCHMITZ (1996)<sup>7</sup>.

In order to select the most promising technologies for ASSB production, an efficient yet sound evaluation is required. Within strategic technology planning, **technology assessment** typically takes place in a multi-step procedure (HAAG ET AL. 2011, p. 317). This holds particularly true for the evaluation of technology chains, where the effort can rise drastically with the amount of technologies taken into consideration (FALLBÖHMER 2000, 48f.). A preselection of alternatives

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<sup>5</sup> While further activities, such as technology scanning, technology monitoring, and technology scouting, mainly focus on the identification of emerging trends and technologies, technology identification also takes into account established technologies (GREITEMANN ET AL. 2016).

<sup>6</sup> For an overview of further approaches for technology identification, please refer to GREITEMANN (2016, 16ff.). English readers are referred to GREITEMANN ET AL. (2015), GREITEMANN ET AL. (2016), and GREITEMANN ET AL. (2017).

<sup>7</sup> Further approaches for strategic technology planning can be found, for instance, in BURGSTAHLER (1997) and SCHÖNING (2006).

is usually based on qualitative means and can, for instance, be carried out by the definition of exclusion criteria (SCHMITZ 1996, 35f.), by technology portfolios (MARKOWITZ 1952; PFEIFFER ET AL. 1982), or using the Analytic Hierarchy Process (AHP)<sup>8</sup> (SAATY 1977; SAATY 1990). The remaining options are then further evaluated with regards to economic aspects (SCHÖNING 2006). Typical methods include the net present value method, total cost of ownership approaches, amortization calculations, or break-even analyses (HAAG ET AL. 2011, p. 320). A large variety of different quantitative and qualitative criteria for technology assessment has been suggested in scientific literature<sup>9</sup>. With regards to the requirements deduced in section 3.1, the focus of this thesis will be on the technical feasibility<sup>10</sup>, technology maturity<sup>11</sup>, and cost<sup>12</sup>.

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<sup>8</sup> The *Analytic Hierarchy Process* is a method in which a defined number of qualitative criteria are quantified by the subjective assessment of experts and thus made comparable with each other (SAATY 1990).

<sup>9</sup> Typical criteria are the estimated achievable technology performance, expected costs and benefits, opportunities and risks, (strategic) relevance for the company, and implementation effort (HAAG ET AL. 2011, p. 315). MARTINI (1995, p. 35) lists, amongst others, the technology performance, capital intensity, flexibility, and level of automation. Similar criteria can be found, e.g. in KRÖLL (2007, 93ff.) and HEIMES (2014, 125ff.), who address the cost, quality, time, flexibility, and maturity. SCHINDLER (2015) also takes into account the technology potential which was categorized by HOFER ET AL. (2019) based on economic, technical, and strategic aspects. Other authors, such as SCHÖNING (2006) and MÜLLER (2008), focus on a purely monetary assessment.

<sup>10</sup> *Technical feasibility* describes the basic producibility of the product (KLOCKE ET AL. 2000). Based on considerations by ESAWI & ASHBY (1999) and FALLBÖHMER (2000), SCHINDLER (2015) suggests to assess the technical feasibility based on materials properties (chemical, mechanical, physical, electrical, and magnetic properties), product properties (geometric dimensions, form elements, tolerances, surface features, etc.), and production properties (process forces, planned quantity ranges, throughput, degree of automation, etc.).

<sup>11</sup> Based on the original Technology Readiness Level (TRL) approach by the National Aeronautics and Space Administration (NASA) (MANKINS 1995) and the approach for technology maturity assessment by BROUSSEAU ET AL. (2009), REINHART & SCHINDLER (2010) developed a method to systematically estimate the maturity of production technologies. This approach classifies technologies according to seven TRLs, i.e. basic technology research, feasibility studies, technology development, technology demonstration, resource integration, integration into the production environment, and serial production. Further approaches based on this model were presented, e.g. by FISCHBACH (2013), PETERS (2015), or VELLA ET AL. (2018).

<sup>12</sup> Methods for cost-based assessment during strategic technology planning are widely available in literature, e.g. ESAWI & ASHBY (1999), SCHÖNING (2006), and MÜLLER (2008). In a battery production context, in particular the material, personnel, energy, maintenance, and area cost, as well as economic depreciation and calculatory interest need to be taken into account (SCHÜNEMANN 2016, 35ff.). For more information, please refer to NEBL ET AL. (2006), HORSCH (2015), or MUMM (2015).

#### 3.2.2 Technology Chain Planning

In a technology chain, the output of one technology represents the input of the following technology (DENKENA ET AL. 2011). Individual parts, product components, or products are changed step by step via intermediate states to the final state (DENKENA ET AL. 2011; REINHART & SCHINDLER 2012). Technology chains are independent of manufacturing resources and do not consider secondary processes, such as storage, handling, transport, or quality control (KLOCKE ET AL. 2000). Technology chain planning serves as input for the planning of manufacturing sequences (figure 3.3), which describe a combination of manufacturing resources and also take into account non-value-adding steps (KLOCKE ET AL. 2000). Finally, the production layout can be planned based on the manufacturing sequence (REINHART & SCHINDLER 2012). The following section serves to give an overview of corresponding approaches with relevance for strategic planning of technology chains for ASSB production.

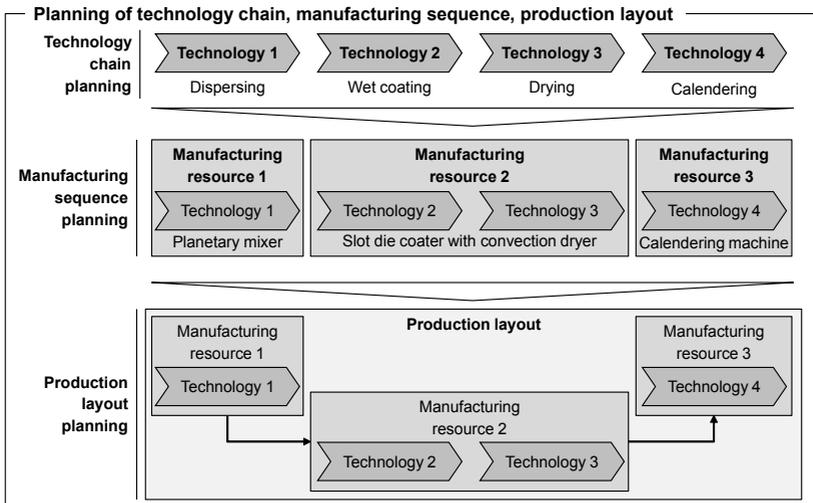


Figure 3.3: Schematic illustration of the framework and level of detail for planning of technology chains, manufacturing sequences, and the production layout. Adapted from REINHART & SCHINDLER (2012).

### 3.3 State of the Art

In this section, the state of the art in strategic planning of technology chains will be analyzed with regards to the underlying research problem. Although there is a large variety of approaches dealing with the evaluation of technology chains from different perspectives<sup>13</sup>, only few of them actually focus on the configuration and assessment of alternative technology chains (or manufacturing sequences) within strategic technology planning. Hence, the focus is on approaches for generation and evaluation of technology chain alternatives in an early production planning phase and on battery production cost modeling.

#### 3.3.1 Technology Chain Generation

Based on the considerations in section 3.1, a selection of potentially suitable approaches for technology chain generation will be briefly summarized and critically reflected with regard to the applicability for ASSB production. An overview is given in figure 3.4, which serves to illustrate different procedures for generation or configuration of technology chains.

FALLBÖHMER (2000) developed a method for the generation of technology chains in early phases of product development<sup>14</sup>. The method starts with a product-technology assignment. Technology-specific characteristics are integrated into a schematically structured technology data model which is based on a classification according to DIN 8580. Technologies are preselected based on the technical feasibility, before alternative technology chains are generated under consideration of interactions between adjacent technologies. Once a core technology<sup>15</sup> has been identified, supporting technologies are assigned with regards to deficits of the core technology to achieve the final product state (figure 3.4, upper image). Intermediate component states are considered which indicate

<sup>13</sup> Examples are, amongst others, the quality-oriented evaluation and optimization approaches by EICHGRÜN (2003), SCHÄFER (2003), DENKENA ET AL. (2011), WESTERMEIER (2016), etc. These are, however, not within the scope of this thesis.

<sup>14</sup> A brief summary of the method by FALLBÖHMER (2000) in English is given in KLOCKE ET AL. (2000). Based on the works by FALLBÖHMER (2000), a method for planning of manufacturing sequences was suggested by TROMMER (2001) (cf. figure 3.3).

<sup>15</sup> A *core technology*, in this context, is a technology which significantly influences the up- and downstream technologies in a technology chain. Note that the term has been ambiguously defined in literature. Other authors, such as MILBERG & MÜLLER (2007), refer to core technologies as value adding technologies (primary technologies).

### 3 Strategic Planning of Technology Chains

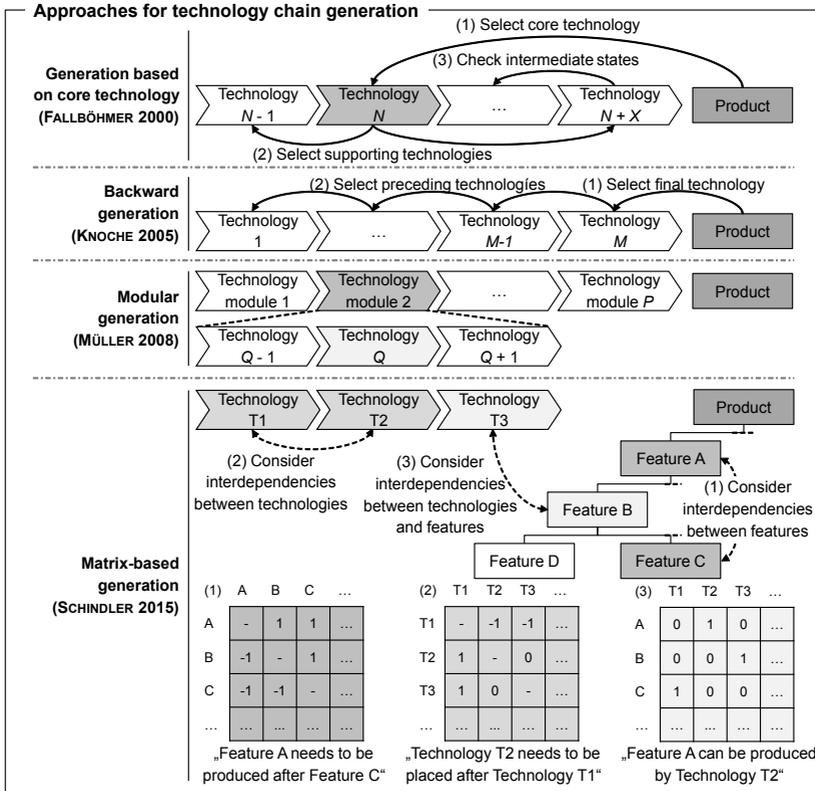


Figure 3.4: Selected approaches for generation of technology chains with relevance for this thesis. Adapted from FALLBÖHMER (2000, pp. 60, 101), KNOCHE (2005, pp. 78, 139), MÜLLER (2008, p. 131), and SCHINDLER (2015, pp. 92, 98).

the need for further supporting technologies within the technology chain. In the last step, newly identified technologies can be integrated. The systematic procedure for technology chain generation by FALLBÖHMER (2000) provides a good foundation for the underlying research problem. However, the routine for technology chain generation will need to be specifically adapted for ASSB production, in particular since also process and assembly technologies need to be considered (cf. section 1.2). Furthermore, the approach is only applicable for

single-component products and is likely to become unhandy for a large number of production technologies.

The generic model for the description of manufacturing technologies by KNOCHE (2005) aims at structuring and linking of technologies. In the first partial model, technologies are classified according to DIN 8580 in a technology tree, whereby machines and specific tools can be assigned. Associated parameters are inherited within this hierarchical structure. The second sub-model describes the applicability of technologies. For each technology, the input and output states are defined, representing the respective component states and acting as so-called features. Each of these features is described by the geometry, material properties, dimensions, and accuracies. In a third model, input and output states are linked and a model to specify the process parameters is built. The fourth partial model is dedicated to the generation of technology chains, where technologies are successively selected in a backward generation approach from the final product state towards the input state(s) (figure 3.4, upper middle image). In addition to the interactions between directly successive technologies, the interactions between non-adjacent technologies are also investigated. The systematic product and technology modeling approach by KNOCHE (2005) lays a sound foundation to cope with the complexity of the underlying research problem. However, the backward generation of technology chains seems hardly applicable for a product with multiple components such as the ASSB.

MÜLLER (2008) presents a further approach for the generation (and evaluation) of technology chain alternatives<sup>16</sup>. For the allocation of technologies to product components, product classes are subdivided hierarchically and company-specific primary technologies (exclusively value-adding technologies) are classified and specified using a technology model<sup>17</sup>. Multi-index matrices are used to describe the relations between product and technologies and to consider interactions between production technologies. Preceding and subsequent technologies (secondary technologies) are assigned to each primary technology

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<sup>16</sup> A brief summary in English can be found in MILBERG & MÜLLER (2007). Based on the works of MILBERG & MÜLLER (2007) and MÜLLER (2008), a methodology to generate robust technology chains under consideration of undesired interactions was presented by KLOCKE ET AL. (2017).

<sup>17</sup> Similar to the approaches by FALLBÖHMER (2000) and KNOCHE (2005), manufacturing technologies are classified according to DIN 8580. The generic model for the technological information is based on the feature description by KNOCHE (2005).

### 3 Strategic Planning of Technology Chains

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in order to create a standardized technology module (figure 3.4, lower middle image). The generation of technology chains is based on the procedure by FALLBÖHMER (2000). However, it is expanded to also consider the fabrication and joining of several components. Structural measures, such as elimination or exchange of technologies, are introduced for the analysis and adjustment of the generated technology chains. In principle, an adaption of the approach for generation of technology chain modules by MÜLLER (2008) seems plausible for the underlying research problem. However, an adjustment of the routine for selection of technologies will be required since secondary technologies are not within the scope of this thesis. Furthermore, the modeling logic is quite abstract, potentially resulting in a high implementation effort.

HEIMES (2014) developed a methodology for the selection of manufacturing resources in LIC production<sup>18</sup>. Technology chains and manufacturing sequences are generated in an integrated Quality Function Deployment (QFD)<sup>19</sup> approach based on three modules: In the initiation module, the desired product properties and process characteristics are defined. In the analysis module, alternative manufacturing resources are identified and evaluated using a morphology box for visualization and the AHP for assessment. This is followed by an analysis of the interdependencies between product properties and manufacturing resources, as well as the interactions between different manufacturing resources. In the third module, manufacturing resources are selected and, thus, the technology chain and the manufacturing sequence (cf. figure 3.3) are generated with aid of the morphology box. This is done by comparing the defined process characteristics with the properties of the manufacturing resources taking into account the identified interdependencies. In a final step, the generated technology chain is checked for consistency. As the focus of the approach by HEIMES (2014) is on manufacturing resources, the level of detail exceeds the amount of information currently available for ASSBs. Furthermore, this approach is only applicable for technology chains with similar sequences and, therefore, needs to be individually adapted for different ASSB cell designs. Nonetheless, a simplified

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<sup>18</sup> English readers are referred to KAMPKER ET AL. (2012) and KAMPKER ET AL. (2013).

<sup>19</sup> *Quality Function Deployment* is a matrix-based method to map customer requirements with technical parameters. Multiple matrices can be used to detail the specifications down to the process level (AKAO 1990).

approach adapted towards the evaluation of technology chains seems plausible, especially due to the specific focus on battery production.

The methodology by SCHINDLER (2015) is designed to assist in strategic planning of technology chains<sup>20</sup>. At first, the technology strategy is defined and the product is modeled<sup>21</sup> in order to determine the production task. In a second step, technology alternatives are identified and preselected. Subsequently, alternative technology chains are generated using Design Structure Matrices (DSMs)<sup>22</sup> and Domain Mapping Matrices (DMMs)<sup>23</sup> to map interdependencies between features, between technologies and between technologies and features (figure 3.4, bottom image). The alternative technology chains are evaluated based on a multi-criteria assessment and the technology chain with the highest degree of suitability is selected. Although the approach for strategic evaluation of technology chains by SCHINDLER (2015) addresses many aspects relevant for this thesis, an adaption towards the specific requirements of ASSB production will be necessary. In particular the interactions between different components are not covered and need to be addressed separately.

#### 3.3.2 Battery Production Cost Modeling

Due to the large investment expected for large-scale ASSB production, the final evaluation of technology chains must take place based on economic criteria. Typically, the cost for energy storage systems is compared based on their energy content (i.e. in cost per kWh) rather than cost per piece (SCHÜNEMANN 2016, p. 96). This facilitates the comparison of different battery systems, which is of particular relevance for the underlying work. Therefore, a battery-specific modeling approach<sup>24</sup> is required (SCHÜNEMANN 2016). The following paragraphs serve to give an overview of recent literature (since 2010) for analyzing the

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<sup>20</sup> For a summary in English, please refer to REINHART & SCHINDLER (2010), REINHART ET AL. (2011), and REINHART & SCHINDLER (2012).

<sup>21</sup> Similar to KNOCH (2005), SCHINDLER (2015) also employs a feature-based method.

<sup>22</sup> A *Design Structure Matrix* is a symmetric matrix which can be used to map interdependencies of entities from the same domain (STEWART 1981).

<sup>23</sup> A *Domain Mapping Matrix* is a matrix which can be used to map interdependencies of entities from different domains (MAURER & LINDEMANN 2008).

<sup>24</sup> A generalized, abstract model for economic evaluation of process chain alternatives was presented, for instance, by GÖTZE ET AL. (2013).

### 3 Strategic Planning of Technology Chains

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production cost of LICs. Similar approaches for SOFCs will also be indicated due to the similarity with some ASSB types (cf. section 2.3).

**Top-down models** provide a simple tool for cost estimation due to the minimum amount of input required (SCHÜNEMANN 2016, p. 2). A top-down model for LIB cost estimation has been used, for instance, by BERG ET AL. (2015), where material costs are calculated based on the cell design, while manufacturing costs (labor, overhead, etc.) are added as fixed rates. Top-down models are often used as a basis for predictions based on learning curves<sup>25</sup>, for instance by BERCKMANS ET AL. (2017), to estimate the future cost of LIC with different cell chemistries<sup>26</sup>. An estimation of the material cost for different LICs and ASSBs has been presented by SCHMUCH ET AL. (2018), not taking into account processing and overhead cost. The drawback of this type of calculation is the considerably low level of detail and the high uncertainty of the information to be obtained, which can therefore only serve as a rough estimate of the expected cost (SCHÜNEMANN 2016, 2f.).

More valuable information on the cost evolution along the technology chain can be gained by so-called **bottom-up models**, where the cost occurring in every process step is modeled individually. A bottom-up model has been used, for instance, by BARNETT ET AL. (2011) and REMPEL ET AL. (2013) to calculate the production costs of large-format LICs, taking into account the process steps from electrode manufacturing up to the cell formation and test<sup>27</sup>. Uncertainties are mapped via Monte Carlo Simulation (MCS)<sup>28</sup> to account for sensitivity of the model, with a special focus on different cathode materials and thicknesses. The MCS allows to account for a multi-variable uncertainty, which is of particular

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<sup>25</sup> The *learning curve* or experience curve describes the effect that production costs drop by a certain percentage when doubling the cumulative production output (WRIGHT 1936).

<sup>26</sup> Learning curves have also been fitted using historical data to project the future cost for different energy storage devices, including battery packs and different types of LICs (NYKVIST & NILSSON 2015; SCHMIDT ET AL. 2017). Similar approaches for SOFCs can be found, e.g. in RIVERA-TINOCO ET AL. (2012) and STAFFELL & GREEN (2013).

<sup>27</sup> Similarly, approaches for bottom-up calculation of SOFC production cost have been presented by OTOMO ET AL. (2013), SCATAGLINI ET AL. (2015), SCATAGLINI ET AL. (2017).

<sup>28</sup> The MCS is a method often used to handle uncertainty in complex prediction models. Here, the input parameters are defined by a distribution (density) function, from which a random value is drawn to calculate the output results. By repeating this procedure with a sufficiently large number of runs, histograms can be generated to illustrate the probability distribution of the model output (RUBINSTEIN & KROESE 2017).

interest for the targets of this thesis. However, the input data and the calculation logic of the model are not disclosed, resulting in a lack of transparency.

The Battery Performance and Cost model (BatPaC) by NELSON ET AL. (2011) and GALLAGHER & NELSON (2014) is regarded as one of the most complete works in the field (PATRY ET AL. 2015; SCHÜNEMANN 2016). It is used to analyze an entire battery pack for automobiles including a cell design model for pouch cells as well as a bottom-up cost model for the corresponding technology chain. Proportionality factors are used to take into account economies of scale for varying output quantities. As the BatPaC modeling tool is universally accessible to the public, it has been used by several authors as a starting point for their own (extended or adapted) models or as a reference<sup>29</sup>. The data origin and essential formulae for battery modeling are extensively documented and explained, which facilitates a comprehension of the results and ensures transparency. However, a change in process steps sequence is not intended in the model.

A different model was presented by SCHÜNEMANN (2016), who developed an instrument for forecasting manufacturing costs for large-format LICs. The study's focus is to gain knowledge regarding the specific cost structure of the battery cell production process. Therefore, a specific calculation logic was developed with which material flow and value stream can be mapped quantitatively and qualitatively along the technology chain. The model is fed with data from machine tool suppliers, raw material quotations, field studies, expert knowledge, and literature research, as well as empiricism from a research pilot line. In addition to developing the theoretical model approach, the work also aims to identify concrete recommendations to reduce manufacturing costs. The author uses a reference parametrization to validate the developed calculation model in

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<sup>29</sup> Similar models were, for instance, used by BRODD & HELOU (2013) and CHUNG ET AL. (2016) to compare manufacturing cost for LIB production in different countries, with a focus on wages, logistics, automation level, etc. NELSON ET AL. (2015) investigate to what extent flexible manufacturing plants can produce different types of batteries and, thus, achieve economies of scale. PATRY ET AL. (2015), SAKTI ET AL. (2015), and AHMED ET AL. (2018) investigate the cost of automotive LICs with a special focus on the influence of the cathode properties (e.g. cathode materials, coating thickness/width, or upper cutoff voltage) on the overall cost. Other factors investigated are, for instance, fluctuations of material prices or the number of layers per cell. A simplified model is used by WOOD ET AL. (2015) for investigation of cost reduction potentials during electrode manufacturing and the wetting and formation procedure. CIEZ & WHITACRE (2017) built up a model for comparison of cylindrical and prismatic cells.

### 3 Strategic Planning of Technology Chains

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comparison with BatPaC<sup>30</sup>. The publication covers many aspects that are also relevant for this thesis. The model is very descriptive and the calculations and assumptions are presented transparently. The abstract modeling logic allows for adaptation of process steps and, thus, ensures flexibility towards different technology chain sequences. Unfortunately, in contrast to BatPaC, the model itself is not accessible to the public, and the layout of the cell design reveals significant weaknesses. Although a sensitivity for different parameter sets is presented, uncertainties in the model cannot be modeled explicitly.

#### 3.4 Action Required

Based on the considerations on the state of the art in the previous sections, the following conclusions can be drawn: While several of the established methods in strategic planning of technology chains provide a sound basis to address the underlying research problem, a tailoring towards the specific and formal requirements deduced in section 3.1 is required. With regards to the production task, a detailed ASSB product model applicable for the systematic description of various types of ASSB components and materials is fundamental. This is particularly relevant for the generation of technology chains, where most of the current approaches only focus on manufacturing of single-component products, whereas interdependencies between different components (e.g. SES and composite cathode, cf. section 2.3) are not sufficiently covered. Furthermore, the material selection for ASSBs strongly influences the technology chain sequence (cf. section 2.3), necessitating flexibility in technology chain configuration and evaluation. This holds particularly true for the economic assessment, where a comparison with conventional LIC production is required to analyze whether a competitive production of ASSBs is possible. Hence, in the following chapter, a methodology specifically adapted towards strategic planning of technology chains for the production of large-format ASSBs will be presented.

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<sup>30</sup> The model has been used in further evaluations by SCHÜNEMANN ET AL. (2016) and KWADE ET AL. (2018), e.g. for estimating the potential cost reduction induced by replacing the electrode wet coating with an extrusion process.

## 4 Methodology—Strategic Technology Planning for All-Solid-State Battery Production

Based on the challenges presented in chapter 2 and chapter 3, the methodological procedure for strategic planning of technology chains for industrial production of large-format all-solid-state batteries (ASSBs) will be presented in the following. Section 4.1 serves to give an overview of the methodology, which will be detailed within the four publications summarized in section 4.2.

### 4.1 Overview of the Methodology

The methodology for strategic planning of technology chains for industrial production of large-format ASSBs is based on the procedure developed by SCHINDLER (2015). However, as deduced in section 3.4, it was adjusted towards the specific requirements for ASSB production formulated in section 3.1. An overview of the steps and procedure is given in figure 4.1. The realization of the methodology within the publications **P 1** to **P 4** is also indicated. The structure of the methodology allows to iteratively go through the individual steps to successively refine the results.

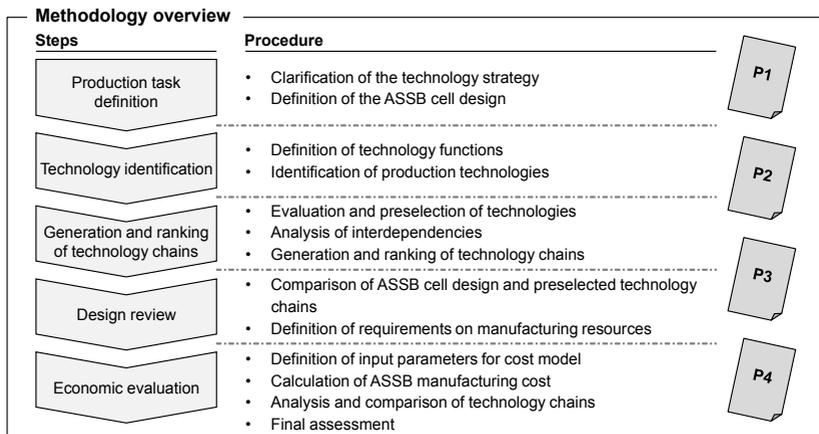


Figure 4.1: Overview of the methodology for strategic planning of technology chains for ASSB production and coarse assignment of the publications **P 1** to **P 4**.

## 4 Methodology—Strategic Technology Planning for All-Solid-State Battery Production

### 4.1.1 Definition of the Production Task

The first step of the methodology is the definition of the production task (figure 4.2). Scope is a clarification of the targeted ASSB product design and the underlying production requirements.

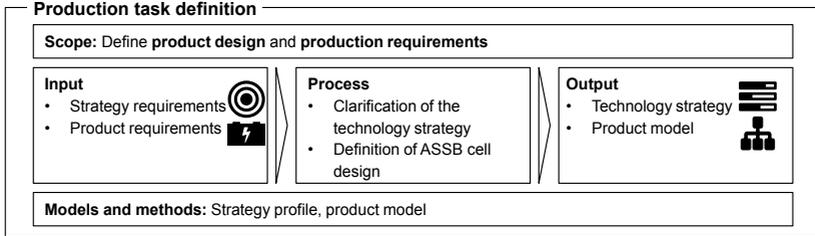


Figure 4.2: Overview of the procedure for definition of the production task.

At first, the technology strategy needs to be defined (cf. section 3.2). This entails in particular a clarification of the underlying objective, such as the planned production output, the targeted markets, timing of market entry, risk affinity, etc. The resulting strategy profile serves as a basis to weight evaluation criteria and to make appropriate decisions for technology selection in the further course of the application.

The targeted ASSB properties are defined with the aid of a hierarchically structured product model, containing relevant information on the planned dimensions, composition, and materials for the ASSB components. A range can be defined for each parameter to take into account the information uncertainty and to allow for product adjustments within the frame of Simultaneous Engineering. Further attributes can be derived by geometrical considerations, for instance by calculating the anode dimensions based on the cathode layer thickness and composition (cf. equation 2.6). Taking into account the solid electrolyte separator (SES) and the current collectors, the number of layers per cell can be derived from the stacking configuration and the housing design (according to DIN 91252). This is a prerequisite to calculate relevant parameters such as the energy density (cf. equation 2.5) and specific energy (cf. equation 2.4) on the cell level. Based on the targeted production output and the ASSB cell design, the

number of ASSBs to be produced can be derived and the amount of material required per ASSB cell and year can be roughly estimated.

### 4.1.2 Technology Identification

The overall goal of the methodology's second step is to build up a pool of technologies potentially suitable for the production of the previously defined ASSB cell design (figure 4.3).

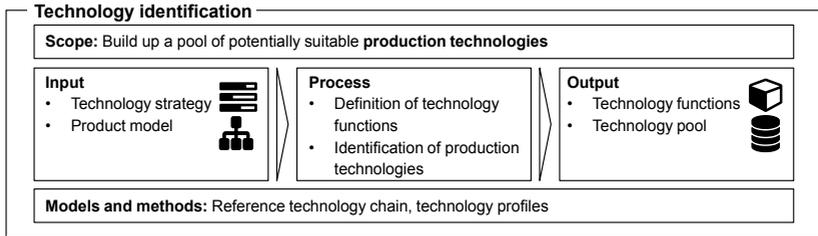


Figure 4.3: Overview of the procedure for technology identification for ASSB production.

Based on the defined ASSB cell design, technology functions<sup>1</sup> are deduced to allow for a solution neutral description of the production task (cf. SCHINDLER (2015, 85f.) and GREITEMANN (2016, 63f.)). Hereby, the fabrication of each ASSB component is described by a chain of technology functions, e.g. “materials mixing”, “layer forming”, and “layer densification”. This can be facilitated by using a reference technology chain (e.g. conventional lithium-ion cell (LIC) production, cf. figure 1.2) and abstracting from discrete production technologies to solution neutral technology functions. Of course, modifications need to be made based on the product structure, for instance integrating the fabrication of the SES layer and omitting the electrolyte filling step.

As suggested by GREITEMANN ET AL. (2016), search fields are derived for each technology function. In order to limit the high expenditure associated with information procurement, these search fields can be prioritized, for instance

<sup>1</sup> A *technology function* according to VDI 2803 serves the purpose of fulfilling the production task and is represented by a noun-verb combination (e.g. “component forming”).

## 4 Methodology—Strategic Technology Planning for All-Solid-State Battery Production

by focusing on components with a high innovation potential (such as the SES layer)<sup>2</sup>. Based on the ASSB cell design and the production output defined within the technology strategy, the required throughput (e.g. the number of sheets to be produced per year) for each technology function can be estimated. The definition of exclusion criteria helps to focus the information acquisition and reject non-suitable technologies already in an early phase (FALLBÖHMER 2000).

Potentially suitable technologies are identified based on the prioritized search fields. A systematic procedure for technology search was presented by GREIT-EMANN ET AL. (2017) (cf. section 3.2). A technology profile helps to structure relevant information on each technology (SCHINDLER 2015, p. 88). The identified technologies are then conglomerated in a technology pool.

### 4.1.3 Generation and Ranking of Technology Chains

In the third step of the methodology, technology chains are generated taking into account the relevant interactions (figure 4.4). Scope is a ranking and prioritization of technology chains for ASSB production.

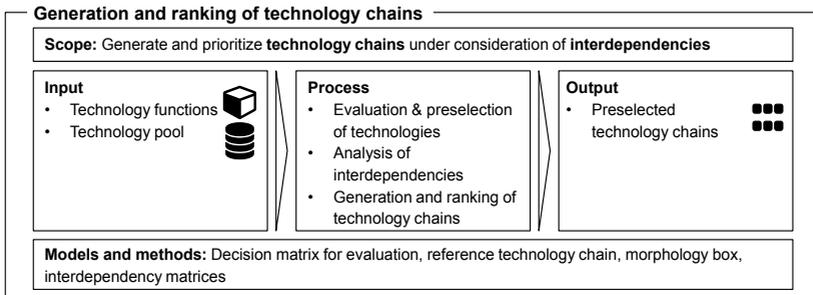


Figure 4.4: Overview of the procedure for generation and ranking of technology chains for ASSB production.

<sup>2</sup> Next to conventional LIC production, promising search fields are production technologies for functional ceramic or crystalline layers in energy conversion or storage, such as SOFCs, multi layer ceramic capacitors (MLCCs), or photovoltaics (PV), but also the polymer and paper industry and related sectors.

In order to reduce the effort for technology chain generation, technologies are preselected based on a multi-criteria assessment: This should take place mainly with regards to technical aspects (cf. section 3.2), such as geometrical constraints or material compatibility (FALLBÖHMER 2000). For a large technology pool, an economic assessment is not advisable at this stage due to the large associated effort (cf. section 3.2). As explained in subsection 3.3.1, the generation of technology chains can be carried out in a similar fashion as suggested by FALLBÖHMER (2000), MÜLLER (2008), SCHINDLER (2015), or a combination of these approaches (cf. figure 3.4). Interactions can be taken into account, for instance, via matrix-based methods (SCHINDLER 2015). Note, however, that also interactions between multiple components need to be considered. A fully-factorial combinatorial analysis of production technologies for each technology function can result in a large number of technology chains<sup>3</sup>. Hence, a pragmatic alternative could be to generate technology chain alternatives by defining one technology chain as a basis and exchanging one technology at a time while taking into account the relevant interactions. The generated technology chains are prioritized, for instance using the average assessment results over the individual technologies in the technology chain (SCHINDLER 2015). Hence, technology chains can be ranked and preselected for further evaluation.

### 4.1.4 Design Review

The fourth step of the methodology is intended to support the Simultaneous Engineering character of the work (figure 4.5). Hence, by analyzing the technical capabilities of the preselected technology chains, the cell design can be specified and requirements for the manufacturing resources can be deduced. This is a prerequisite for the subsequent economic assessment.

As the product characteristics have a significant influence on the overall manufacturing cost, the ASSB cell design needs to be carefully analyzed for each preselected technology chain. Hereby, the product design can be specified by comparing the requirements of the ASSB components (such as targeted layer thickness ranges) with the capabilities of the corresponding technologies. Cell

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<sup>3</sup> Considering, for instance, a chain of ten technology functions with two technology alternatives per technology function, one ends up with  $2^{10} = 1024$  possible technology chains.

## 4 Methodology—Strategic Technology Planning for All-Solid-State Battery Production

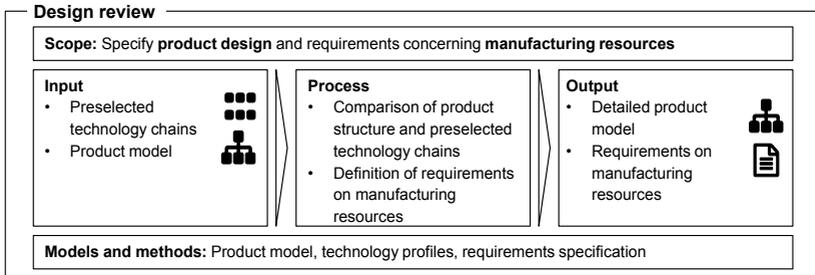


Figure 4.5: Overview of the procedure for design review with regards to product specifications and manufacturing resources for ASSB production.

design adaptations can be made to facilitate a DFMA approach. Furthermore, the specification can also be used to describe requirements on manufacturing resources as defined by KARL & REINHART (2015), for instance environmental conditions (e.g. inert gas atmosphere). The communication with potential technology suppliers is facilitated by conglomerating this information in a requirements specification document, where relevant characteristics (such as targeted layer geometries, throughput, atmospheric requirements, etc.) are summarized. The definition of these characteristics helps to gather specific information on manufacturing resources and, thus, serves as an input for the subsequent economic assessment.

### 4.1.5 Economic Assessment and Final Evaluation

After selecting the most promising technology chains and specifying the ASSB cell design, an economic assessment must take place (figure 4.6). Scope is to estimate the production cost for the selected technology chains to allow for a profound comparison and selection.

Although a top-down calculation can give a first indication of the expected production cost, a comparison of different technology chains will require a detailed bottom-up calculation. This can be done with the aid of a cost model adapted from SCHÜNEMANN (2016) towards the specific requirements of ASSB production. Hence, input parameters on the cell design and the manufacturing resources in the respective technology chains need to be gathered, along with general parameters such as wages, energy pricing, etc. By defining the

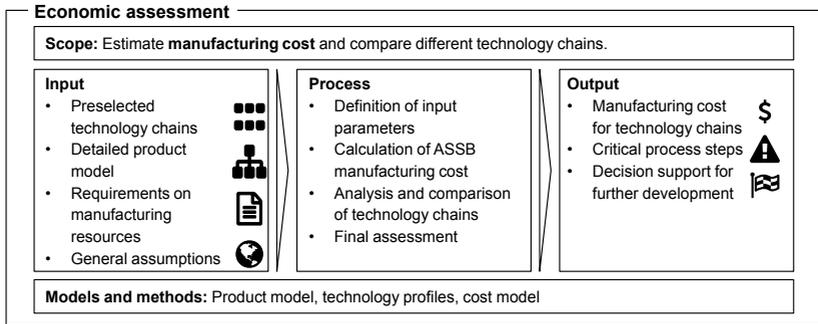


Figure 4.6: Overview of the procedure for the economic assessment of technology chains for ASSB production.

desired production output, the number of machines per process step can be deduced and the amount of input material can be calculated. This allows for a transparent estimation of the ASSB production cost along the whole process chain. Hence, cost critical process steps can be identified and different scenarios and technology chains for ASSB production can be compared. Based on the economic assessment, an iterative procedure can be applied to refine the generated technology chains. This allows for a final selection and facilitates decision support for further development.

## 4.2 Realization of the Methodology

Following the DRM by BLESSING & CHAKRABARTI (2009), the methodology described in the previous section is detailed within a Comprehensive Prescriptive Study in order to provide answers to the research questions presented in subsection 1.3.1. In the following subsections, an overview of the four resulting publications (cf. section 1.4) will be given. The general framework and an ASSB product model are presented in **P 1**. Requirements and challenges for ASSB production are gathered in **P 2**, addressing in particular research question **Q 1**. Research question **Q 2** is mainly subject of **P 3**, where a systematic identification and evaluation of technologies potentially suited for ASSB production is carried through. Finally, a cost model is presented in **P 4** to identify critical factors in ASSB production, as formulated in research question **Q 3**. The hypotheses

## **4 Methodology—Strategic Technology Planning for All-Solid-State Battery Production**

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defined in chapter 2 are mainly addressed within the scenarios investigated in **P 3** and **P 4** and will be finally discussed in subsection 5.2.3.

### **4.2.1 Overview Publication 1**

The paper “Evaluation of technology chains for the production of all-solid-state batteries” (**P 1**) is motivated by the advantages and challenges of ASSB, and the lack of know-how on ASSB production using scalable processes. Hence, the objective of the paper is a method to generate and evaluate technology chains for the production of large-format sulfide- and oxide-based ASSBs. Methods from strategic technology planning, technology identification, and technology assessment were adapted towards the specific requirements for ASSB production.

A product structure was developed which enables a systematic modeling of different types of ASSBs. By abstracting the technology chain for conventional LIC production, a reference technology chain was developed to assist in the identification of production technologies and the generation and evaluation of technology chains for ASSB production. The procedure starts with a selection of potentially suitable production technologies based on a requirements analysis. Hence, technology chains are generated with the help of a morphology box, taking into account the relevant interdependencies. Finally, these technology chains are evaluated based on the technology maturity and economic criteria. The applicability of the method was shown for an exemplary technology chain, where the technology maturity of a tape casting technology for fabrication of a composite cathode was estimated. After a summary of the developed method, the paper concludes that further research will be required to comprehensively analyze the different scenarios for ASSB production and the respective challenges.

### **4.2.2 Overview Publication 2**

Based on the considerations of **P 1**, the paper “All-solid-state lithium-ion and lithium metal batteries – paving the way to large-scale production” (**P 2**) states that a scale-up from laboratory research to industrial mass production will be required for ASSBs to become competitive with conventional lithium-ion

battery (LIB). However, a direct transfer of manufacturing approaches from laboratory scale is in many cases not possible. The scope of the paper is therefore an investigation of challenges and requirements for ASSBs from a production engineering perspective, and to develop possible scenarios for ASSB fabrication. The paper summarizes the results of an empirical study based on expert interviews: A dynamic questionnaire was used to gather challenges for production and to define an ASSB cell design for the subsequent morphological analysis. Here, the experts were asked to generate technology chains and fill a corresponding morphology box to analyze the respective production parameters.

Based on the expert interviews, requirements and challenges for different material combinations and cell designs were identified (cf. **Q 1**) and production scenarios were developed (cf. **Q 2**): For both SES and composite cathode<sup>4</sup> fabrication, a wet chemical processing route or a high-viscosity processing route were identified, while for the SES also powder or vapor-based processing seem plausible (cf. **H 3**). For the lithium anode, foil processing, melt processing, vapor based processing, or electrochemical plating can be applied (cf. **H 5**). The process steps for cell assembly can only be roughly outlined. Single-sheet stacking seems to be most plausible due to the limited bending stiffness of the compacted sheets. Advantages and disadvantages of the respective processing routes are summarized, before giving a comparison with conventional LIC production: For a sulfide-based ASSB, an adaption of established process steps could be feasible if the required production environment (cf. **H 2**) and low porosities can be provided. In contrast, the production of an oxide-based ASSB significantly differs due to the expected high temperature sintering step (cf. **H 1** & **H 4**). The paper concludes with implications and recommendation for stakeholders, such as battery cell producers, chemical industry, and machine engineering. An outlook is given on the next steps required for upscaling, such as experiments on manufacturing and assembly of ASSB components.

### 4.2.3 Overview Publication 3

The publication “Prospects on production technologies and manufacturing costs of oxide-based all-solid-state lithium batteries” (**P 3**) is motivated by the

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<sup>4</sup> Similar considerations apply for a composite anode, e.g. with graphite or LTO.

## 4 Methodology—Strategic Technology Planning for All-Solid-State Battery Production

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issues for oxide-based ASSBs due to the interfacial challenges and the expected necessity for a high temperature sintering step. As deduced in **P 2**, this hinders a transfer from established technologies in conventional LIC production. Therefore, different processing routes are required, for instance based on ceramic manufacturing technologies. Hence, the objective of the paper is the identification and evaluation of production technologies for oxide-based ASSBs. Methods applied in strategic technology planning were adapted to suit the underlying research problem: Hence, a systematic technology identification was carried through for the fabrication of oxide ceramic layers. Exclusion criteria were defined for a preselection of technologies, followed by a rough evaluation of material, product, and production related criteria using the AHP. Subsequently, the most promising technologies were assessed with regards to technology maturity. Here, a MCS was carried through based on the method introduced by REINHART & SCHINDLER (2012). By combining data from conventional LIB and SOFC production, learning rates were applied to enable a top-down cost calculation for an exemplary technology chain.

Based on an analysis of requirements and cell designs for oxide-based ASSBs, potentially suitable manufacturing technologies were systematically identified (cf. **Q 2**). Search fields are in particular the fabrication of functional ceramic layers in energy conversion or storage, such as SOFC and MLCC production. The identified layer fabrication technologies were preselected based on the material compatibility (thermal stability) and error rate for fabrication of layers with the previously defined layer geometries. Thereby, most plasma- and flame-based spray coating and vapor deposition technologies were excluded (cf. **H 3**). The AHP reveals that established layer fabrication technologies, such as tape casting and screen printing, fulfill the production related criteria best. However, a high temperature sintering step is required as post-treatment (cf. **H 1**). In contrast, the ADM fulfills the product related criteria best, in particular the fabrication of dense, ion-conducting layers without sintering. However, the subsequent TRL assessment reveals a very low maturity of the ADM, indicating a high development effort in order for the technology to become capable for ASSB mass production (cf. **H 3**). Hence, the exemplary technology chain used for the top-down cost calculation is based on tape casting (for the SES) and screen printing (for the cathode) with two sintering steps, similar to SOFC production (cf. **H 4**). The analyzed scenarios with different cathode materials and thicknesses indi-

cate that ASSB cost could potentially become competitive with conventional LIBs by economies of scale. The paper reveals that the challenges at the interfaces in oxide-based ASSBs are directly linked to the respective production technologies. Hence, Simultaneous Engineering will be required to enable the cost competitive fabrication of oxide-based ASSBs with high energy and power density and long cycle life. This could also be a potential for equipment manufacturers and related companies with ceramics processing know-how. Finally, the publication concludes that a detailed bottom-up calculation will be required to identify critical process steps in ASSB production.

### 4.2.4 Overview Publication 4

The paper “Solid versus Liquid—A Bottom-Up Calculation Model to Analyze the Manufacturing Cost of Future High-Energy Batteries” (P 4) is motivated by the high uncertainty and the risk associated with an investment in ASSB production. As outlined in P 3, a detailed assessment of the production steps is required in order to identify critical processes (cf. Q 3). Hence, the objective of the publication is to estimate the production cost for ASSB production along the entire process chain for different scenarios. For this purpose, a bottom-up calculation model was built up based on the calculation logic developed by SCHÜNEMANN (2016). The input data was empirically gathered based on literature, expert interviews, and technology supplier quotations. In order to cope with the information uncertainty, input parameters can be systematically varied and a MCS was implemented.

By comparing a conventional LIB (graphite or Si/C anode) with a sulfide-based ASSB (graphite or lithium anode), the costs for components manufacturing, cell assembly, and formation/testing were assessed, taking into account in particular also the dry room and inert gas housing cost (cf. H 2). While an ASSB with graphite anode will result in the highest overall cost, production of a ASSB with lithium metal anode could even be cheaper than a conventional LIB with Si/C anode. An analysis of the lithium foil production process by extrusion and calendaring reveals that in-house production will be beneficial compared to purchasing the foil externally (cf. H 5). Furthermore, different ASSB cell designs and SE materials were investigated: While a bipolar stacking enables a higher voltage and better packaging utilization, processing cost can be higher than for a

## 4 Methodology—Strategic Technology Planning for All-Solid-State Battery Production

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conventional parallel stacking design. The higher density of the oxide-based SE and the high-temperature sintering step result in higher material and processing cost in comparison to the sulfide-based ASSB (cf. **H 1** & **H 6**). This is confirmed by a sensitivity analysis where all input parameters were systematically varied and a MCS with a variation of input parameters for inert gas environment (for sulfide-based SE) and sintering (for oxide-based SE). In order to circumvent the highly critical sintering step, the ADM was investigated as an alternative for oxide-based ASSBs. Although this would enable an improved cell design and different SE material combinations, deposition rates are currently far from economic application of the technology (cf. **H 3**). The paper concludes that a detailed cost assessment of the investigated scenarios can help to mitigate the risks associated with a scale-up of ASSB production. However, the underlying assumptions need to be experimentally validated, which necessitates a transfer from the laboratory scale towards pilot lines.

### 4.3 Summary

In summary, a methodology for strategic planning of technology chains for production of large-format ASSBs was developed and realized within a Comprehensive Prescriptive Study. By analyzing challenges, requirements, and possible production scenarios for ASSB production from both a technical and an economic perspective, the four publications significantly contribute to the knowledge base in the research field. In the following chapter, the methodology shall be applied in an industrial context and evaluated with regards to the requirements defined in chapter 3. In this way, the hypotheses formulated in chapter 2 can be critically discussed and answers to the research questions defined in subsection 1.3.1 can be derived.

## **5 Discussion—Application and Evaluation**

In this chapter, the results of a selected application example will be presented which serves to critically reflect the methodology developed within the previous chapter: In section 5.1, the applicability of the methodology will be verified within an industrial context. Subsequently, in section 5.2, the methodology will be assessed with regards to the benefit-to-cost ratio and the fulfillment of the requirements deduced in chapter 3. Furthermore, the hypotheses derived within chapter 2 will be discussed to conclude on the research questions formulated in chapter 1.

### **5.1 Application**

This section serves to illustrate the application of the methodology using a selected example. After a brief description of the use case scenario in subsection 5.1.1, the core findings of the application will be summarized within subsection 5.1.2.

#### **5.1.1 Application Example—Battery Cell Manufacturer**

In order to verify the applicability of the developed methodological procedure within an industrial context, the methodology for strategic technology planning of ASSB production was applied together with an international battery cell manufacturer. The company is a market leader for various types of batteries in niche applications from individual battery cells up to large battery storage systems. However, the company expects that with a disruptive battery technology such as the ASSB, it could also enter the competitive automotive market. Hence, an internal strategy team had been set up to identify technologies and processing routes for ASSB production that match the company strategy profile and competencies. In a bilateral research and development project, the methodology was carried through building on the preceding work of the strategy team.

### 5.1.2 Results

The following subsection summarizes the results of the project. In order not to disclose any confidential information, the results were abstracted and company specific information was blurred.

#### Definition of the Production Task

In order to allow for a rough categorization of the company's technology strategy, a questionnaire was set up to gather information on targeted markets, quantities to be produced, the planned time horizon, risk affinity, etc. With regards to the internal competencies of the company, the strategic focus had been set on polymer-based, sulfide-based, or hybrid SEs (cf. subsection 2.2.3). Based on the product model presented in **P 1**, the targeted dimensions and composition of the ASSB components (electrodes, SES, current collectors, housing) were defined for the different material systems<sup>1</sup>. To account for the uncertainty associated with ASSB product design (cf. section 2.4), values were given on a three-point scale (minimum, most likely, maximum). Hence, important characteristics such as the energy density and specific energy could be calculated and compared for the different cell designs.

#### Technology Identification

Based on the results published in **P 2** and **P 3**, the company's strategy team had already started to identify possible production technologies. As suggested in **P 2**, the technologies had been clustered in a morphology box for the different technology functions. The focus was on manufacturing of the ASSB core components (cathode, SES and anode) due to the great uncertainty associated with ASSB cell assembly. Within the project, the technology pool was checked for consistency and extended by further technology functions and production technologies. The final technology pool consisted of 33 technologies in total with a focus on components manufacturing from materials mixing to layer fabrication and post-treatment. In order to allow for a systematic assessment

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<sup>1</sup> The abstract ASSB product model also allowed for a simple adaption towards polymer-based SE.

and preselection according to P 3, technology profiles were prepared for each technology (cf. SCHINDLER (2015, p. 88)).

## Generation of Technology Chains

The technology pool served as input for a workshop held by the author together with the strategy team and selected company internal experts. Objective of the workshop was to prioritize evaluation criteria in accordance with the technology strategy and to assess and preselect the identified technologies using the AHP. Based on considerations by HOFER ET AL. (2019), criteria were clustered according to technical, strategic, and economic aspects: In particular the technical criteria (cf. P 3) were adapted for each technology function, as exemplarily depicted in figure 5.1 for mixing, coating, and densification technologies<sup>2</sup>. Strategic criteria entail, amongst others, the technology maturity, company-specific know-how, technology suppliers, etc.

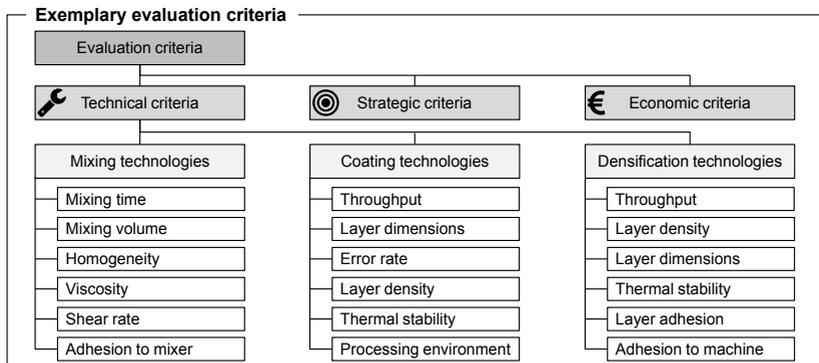


Figure 5.1: Categorization for evaluation criteria based on HOFER ET AL. (2019) and exemplary technical criteria for mixing, coating, and densification technologies.

<sup>2</sup> Although only individual technologies are assessed at this stage, possible interactions with preceding or subsequent technologies need to be taken into account for the evaluation: For instance, the layer density for most slurry-based coating technologies is typically very low (approx. 50 % porosity). Nevertheless, when considering a subsequent densification process, close-to-zero porosities can be achieved (HITZ ET AL. 2019). Hence, rather the effort for densification during post-treatment should be considered when assessing the respective coating technology.

## 5 Discussion—Application and Evaluation

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The workshop consisted of two parts: At first, the technical and strategic evaluation criteria were ranked with aid of a pairwise comparison (cf. **P 3**)<sup>3</sup>. The second part of the workshop was dedicated to assess the identified technologies with regards to criteria fulfillment. Therefore, a DMM was created and filled for each technology function. The results were evaluated and visualized as a portfolio: Here, both technical and strategic criteria were summarized on one axis each, as exemplarily depicted in figure 5.2 (image left) for SES materials mixing<sup>4</sup>. Unsurprisingly, high strategical scores are achieved by technologies applied in conventional LIC production, such as planetary mixing (cf. section 1.2). However, also alternative technologies achieve good ratings, for instance those allowing for continuous mixing and solvent reduction (SCHÜNEMANN ET AL. 2016).

Based on the portfolios, twelve technologies were selected for further assessment. The technology chain for sulfide-based ASSB deduced in **P 2** was deployed as a reference for technology chain generation. Using the technology functions described in **P 2**, further technology chains for the other preselected technologies were generated by a systematic variation of selected technologies (figure 5.2, image right). Taking into account the relevant interactions (cf. **P 2**), one technology was exchanged at a time and all other technologies were kept constant if technically feasible (cf. FALLBÖHMER (2000)). Hence, eight different technology chains were selected for further assessment.

### Design Review

Based on the generated technology chains, the battery cell design was refined with regards to the selected technologies (FALLBÖHMER 2000). For instance, the lithium anode layer thickness was specified from the previously defined thickness range depending on the technical capability of the layer fabrication technology: While thinning down lithium foil by rolling is physically limited to approximately 20  $\mu\text{m}$  thickness, much thinner layers could be achieved by

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<sup>3</sup> As suggested in **P 1**, economic parameters were not considered at this stage due to the high effort associated with the large number of technologies to be assessed.

<sup>4</sup> The diagram was abstracted due to confidentiality reasons.

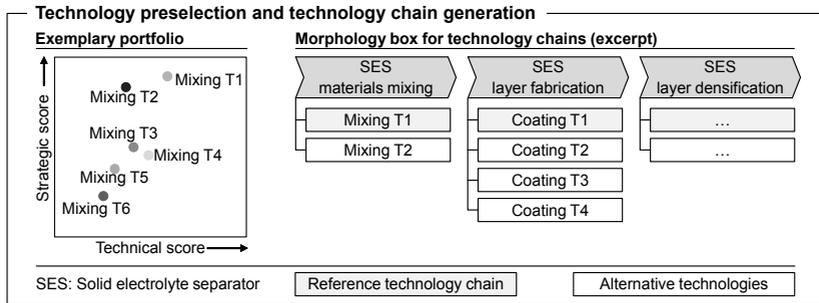


Figure 5.2: Exemplary portfolio for SES materials mixing and subsequent generation of technology chains based on a morphology box and a reference technology chain.

alternative methods (cf. P 2). Hence, the cell design was individually adapted depending on the processing route.

## Economic Assessment

The reference technology chain for sulfide-based ASSBs (P 2) served as a baseline scenario for the economic assessment. Where suitable, input data was taken from P 4 and adapted towards the specific use case scenario. Based on a requirements specification on manufacturing resources (e.g. materials to be processed, desired throughput, etc.), supplier quotations were gathered for all technologies not covered in P 4. Exemplary results for the cost assessment are summarized in figure 5.3.

Figure 5.3 (image left) shows the overall cell cost breakdown per kWh for different composite cathode processing routes: The left bar corresponds to the reference scenario. For the other two scenarios, the mixing technology was exchanged. A solvent-free processing route was assumed for the right bar. The latter reveals a savings potential of 8 % compared to the other two examples, as material cost (no solvent) and energy consumption (no solvent evaporation) are significantly lower. This is in good agreement with calculations by WOOD ET AL. (2015) and SCHÜNEMANN ET AL. (2016) for a solvent reduction in conventional LIC production.

## 5 Discussion—Application and Evaluation

Figure 5.3 (image right) illustrates the potentials of replacing the conventional mechanical thinning step for lithium anode production by an alternative process: A reduction of the lithium film thickness would allow to significantly enhance the energy density (left axis)<sup>5</sup>, for instance from approximately 700 Wh/L for 20  $\mu\text{m}$  to approximately 800 Wh/L for 5  $\mu\text{m}$ . A higher energy density also leads to a reduction in manufacturing cost per kWh (right axis) for both processing routes. Fabrication of a 20  $\mu\text{m}$  thick film using the alternative technology will result in higher costs than for the reference. However, the throughput for the alternative processing route will be drastically increased for thinner layers and, thus, less machines will be required. This results in a stronger cost decrease for thinner layers in comparison to the conventional rolling procedure.

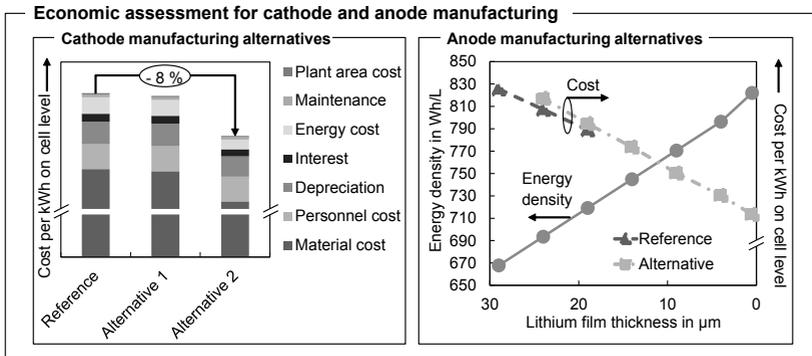


Figure 5.3: Cost assessment for different cathode and anode processing routes. Image left: Overall cell cost breakdown for the reference scenario and two alternative processing routes. Image right: Energy density (circles, left axis) for different lithium foil thicknesses and resulting cell cost (right axis) for the reference (triangles) and an alternative technology (squares).

### Final Selection

In an iterative procedure, the technology chains with the best economic, technical, and strategic score were generated, highlighted, and compared with

<sup>5</sup> Note that the amount of lithium needs to be carefully balanced to account for losses during cycling (GENOVESE ET AL. 2018).

regards to the initial reference (figure 5.4) with the aid of a Strengths, Weaknesses, Opportunities, Threats (SWOT) analysis (HILL & WESTBROOK 1997)<sup>6</sup>. Unsurprisingly, a cost focus leads to the best economic result ( $-8\%$  compared to the reference), while a technical focus is expected to result in higher costs ( $+13\%$ ) but also higher product quality, for instance by a more homogeneous densification. The strategic focus allows for a fast scale-up along with a cost reduction potential. Based on these results, recommendations for the next steps towards ASSB production were given.

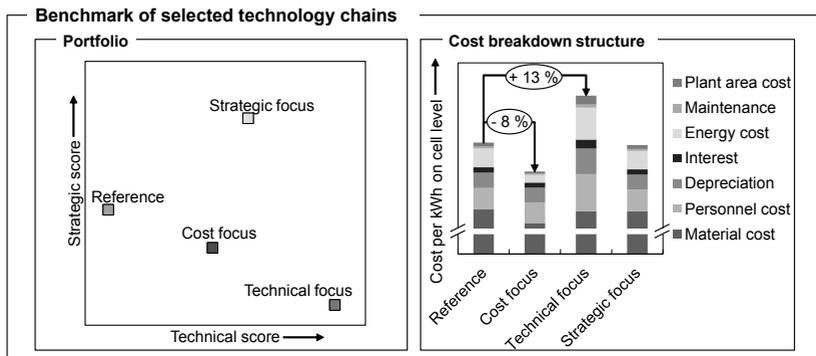


Figure 5.4: Portfolio and cost breakdown structure for the selected technology chains.

### 5.1.3 Critical Reflection

In summary, the applicability of the methodology was successfully verified together with an international battery cell manufacturer. This proves that the methodology can be applied in an industrial context. While some of the results could not be disclosed due to confidentiality reasons, the given figures and examples significantly enhance the understanding of the underlying methods and facilitate a graphical visualization.

Within the application example, the method by FALLBÖHMER (2000) was adapted for technology chain generation. Thus, only a selected number of technology chains were generated, which is in contrast to the matrix-based

<sup>6</sup> The final selection cannot be disclosed due to confidentiality reasons.

## 5 Discussion—Application and Evaluation

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procedure suggested by SCHINDLER (2015). In order to check the selected approach for plausibility, technology chains were also generated and ranked automatically with regards to the underlying combinatorics using Microsoft® Excel®. However, the automated ranking revealed that deviations from the manually generated technology chains were mainly due to supporting technologies (cf. figure 3.4) with little relevance for the underlying context, such as laser cutting vs. punching. Therefore, the suggested procedure can be justified, in particular with regards to a reasonable benefit-to-cost ratio (cf. section 3.1).

The biggest challenge that occurred within the application example was to gather economic parameters, especially for technologies with a low degree of maturity. While an economic assessment is in many cases possible for laboratory equipment where quotations can be obtained from corresponding suppliers, a simple transfer towards industrial application has turned out to be challenging due to a lack of knowledge. For instance, physical vapor deposition (PVD) is often used for thin-film ASSBs to fabricate layers in the submicron range; however, no large-scale industrial application for deposition with several  $\mu\text{m}$  thickness has been known so far for ASSB materials. A possible solution could be a transfer from other industries: Similar processes can be found, for instance, in the PV industry or related sectors where roll-to-roll deposition of organic materials or aluminum has been successfully implemented for mass production. By considering the adapted boundary conditions, such as materials characteristics and safety constraints, a rough estimation of the investment, energy consumption, machine area, etc. can be made. Nonetheless, the effort associated with gathering economic parameters must not be underestimated. For the scenarios described in **P 4**, the given input parameters can be adjusted to the specific use case. This helps to significantly reduce the expenditure for information search during economic assessment.

### 5.2 Evaluation

The application example confirms that the methodology can be applied in an industrial context. In the following, the methodology will be evaluated with regards to the benefit-to-cost ratio and the overall requirements fulfillment. Furthermore, the hypotheses derived in chapter 2 will be discussed and confirmed or rejected if possible.

### 5.2.1 Benefit-to-Cost Ratio

As described in section 3.1, the effort for carrying through the methodology for strategic planning of technology chains for ASSB production must be justified with regards to the resulting benefits. Hence, an estimation of the associated cost will be given based on the five steps of the methodology. Hereby, one staff day (SD) corresponds to 8 h.

At first, the production task needs to be clarified. The effort for defining the technology strategy within a team workshop is estimated to be 4 SD. Furthermore, the ASSB cell design must be specified. Most stakeholders in the field of LIC production already have implemented specific LIB product models, for instance as Microsoft® Excel® calculation tool. However, an adaption towards the ASSB product model presented in **P 1** will require a reprogramming effort. With the aid of equation 2.6 and the input parameters given in **P 3** and **P 4**, this is estimated at 1 SD. Specifying the targeted ASSB cell design and defining the respective ranges is done in a team workshop, for instance together with the product development division (0.75 SD). For technology identification, technology functions (cf. **P 2**) and search fields need to be defined (1.5 SD). The effort for information search and technology profile preparation scales with the number of technologies and is estimated at 0.25 SD per technology<sup>7</sup>. For the generation of technology chains, technologies are evaluated and preselected based on a multi-criteria assessment within a team workshop. The preparation of the input matrices (cf. **P 3**) for the workshop is estimated at 0.5 SD. During the workshop, evaluation criteria are weighted (1.25 SD) and technologies are assessed (0.25 h per technology)<sup>8</sup>. The results are summarized in a portfolio (0.5 SD for preparation). Hence, technology chains can be generated by defining a reference technology chain (0.25 SD) and systematically exchanging one technology at a time (in average 0.5 h per technology chain)<sup>9</sup>. Based on the generated technology chains, the ASSB product specification is refined in a

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<sup>7</sup> This corresponds to 8.25 SD for the 33 technologies within the application example.

<sup>8</sup> Hence, an evaluation within a team workshop with five persons corresponds to 5.16 SD for 33 technologies.

<sup>9</sup> Within the application example, twelve technologies were preselected and eight technology chains were generated, corresponding to 0.5 SD. In contrast, the effort for programming a tool that allows for a combinatorial generation of technology chains while taking into account the relevant interactions is expected to account for at least 5 SD.

## 5 Discussion—Application and Evaluation

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design review workshop. This needs to be done individually for all generated technology chains (in average 0.5 h each)<sup>10</sup>. The effort for gathering information for the economic assessment is estimated at 0.5 SD per technology<sup>11</sup>. This entails the preparation of a requirements specification and the identification of and consultation with suitable technology suppliers. The implementation of a cost calculation tool using the modeling logic presented in P 4 accounts for approximately 10 SD<sup>12</sup>. For each technology chain, the calculation needs to be prepared, carried through, and evaluated (0.25 SD per technology chain)<sup>13</sup>. Finally, 1.25 SD are accounted for a team meeting to communicate the results and discuss further steps.

In summary, the overall effort for carrying through the methodology is estimated at 44.4 SD. Hence, the associated cost at a rate of 1.150 \$ per SD sums up to approximately 51.000 \$. The benefit of applying the methodology for strategic planning of technology chains for ASSB production is illustrated using the following example: Using the technology chain for sulfide-based ASSBs described in P 2 as a reference and assuming a production output of 6 GWh/year and a baseline cost of 100 \$/kWh as calculated in P 4, the annual production costs will sum up to 600 M\$. Hence, the savings potential of 8 % per kWh revealed within the application example would result in a 48 M\$ cost reduction per year compared to the reference described in P 2. Thus, the effort for applying the methodology can be justified.

### 5.2.2 Fulfillment of Requirements

Finally, the methodology shall be assessed with regards to the specific and formal requirements deduced in section 3.1. An overview of the requirements and the degree of fulfillment is given in figure 5.5.

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<sup>10</sup> Considering a workshop with three persons, this corresponds to 1.5 SD for eight technology chains.

<sup>11</sup> This corresponds to 6 SD only for the twelve preselected technologies. In order to reduce the effort for assessing the whole technology chain, it is advisable to use the values from P 4 for the remaining technologies.

<sup>12</sup> This can be done using readily available spreadsheet software such as Microsoft® Excel®.

<sup>13</sup> Hence, 2 SD need to be estimated for eight technology chains.

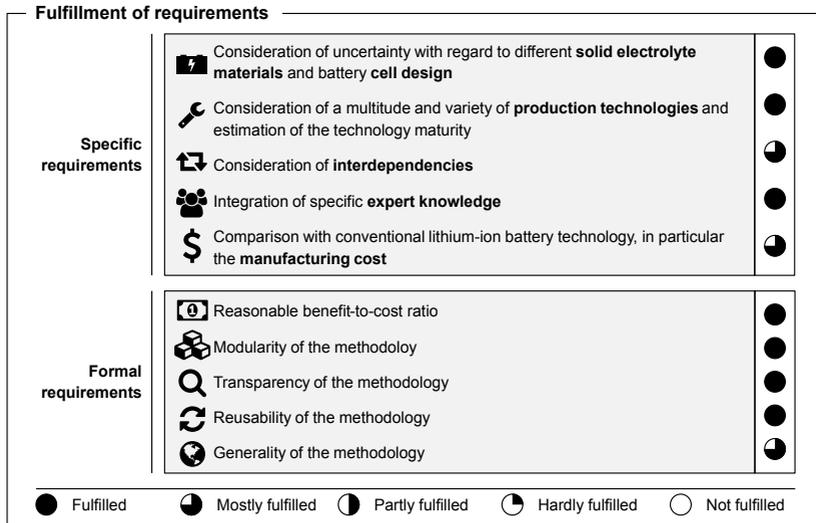


Figure 5.5: Assessment of the methodology with regards to the fulfillment of specific and formal requirements.

## Specific Requirements

Based on the results presented in chapter 4 and the application example, the following conclusions can be drawn on the specific requirements:

- *Consideration of uncertainty with regards to different SE materials and battery cell design:* The abstract ASSB product model developed in **P 1** and **P 4** allows for a structured definition of ASSB properties from materials and components up to the battery cell. Hence, various types of ASSBs with different SE materials can be considered and ranges can be defined to take into account the underlying information uncertainty. By implementing a design review stage within the methodology, an adaption of product properties is enabled to facilitate a DFMA approach. For instance, the layer thickness of an ASSB component can be adjusted based on the technical capability of the selected production technologies. Hence, the methodology allows for a successive reduction of the uncertainty with an increasing knowledge base within the framework of Simultaneous Engineering.

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- *Consideration of a multitude and variety of production technologies and estimation of the technology maturity:* Within this thesis, a large knowledge base was built up with regards to production technologies for ASSB fabrication, from materials mixing to components manufacturing and cell assembly. By providing a multi-step procedure and suitable assessment methods to systematically evaluate and select potentially suitable production technologies, the technology pool can be successively refined and adapted while keeping the expenses for technology assessment within reasonable limits. Hereby, also novel technologies (e.g. the ADM) were systematically assessed, in particular with regards to technical aspects and the technology maturity (cf. P 3).
- *Consideration of interdependencies:* A heuristic procedure was suggested to take into account interdependencies during technology chain generation. The applicability was confirmed within the application example (subsection 5.1.2), in particular with regards to a reasonable benefit-to-cost ratio: Although this procedure will probably not result in an “optimal” technology chain, a direct and transparent comparison is facilitated for all technologies to be regarded. Of course, as suggested by MÜLLER (2008) and SCHINDLER (2015), matrix-based methods could be applied using an automated software tool for combinatorial generation of technology chains. An even more profound selection of technology chains could be provided by a (semi-)quantitative assessment of interdependencies (as suggested by HEIMES (2014)). However, the resulting benefits must be carefully considered with regards to the additional implementation effort, and the high information uncertainty may not allow for a profound assessment.
- *Integration of specific expert knowledge:* The generated knowledge base was built up by numerous interviews and workshops with experts from various disciplines in academia and industry, such as materials science and development, cell manufacturers, machine tool suppliers, and automotive companies. This was supported by different tools for systematic acquisition of information, e.g. the morphological analysis to develop production scenarios and generate technology chains in P 2, or the AHP matrices used to assess the identified technologies in P 3.

- *Comparison with conventional LIB technology, in particular the manufacturing cost:* As discussed within the application example, an economic assessment in early production planning phases is subject to a high uncertainty. Hence, a quantitative cost evaluation for ASSB production is hardly possible, especially if technologies with a low degree of maturity are employed. Nonetheless, based on a systematic comparison with the process chain for conventional LIB production in **P 2**, the cost model developed in **P 4** enables a benchmark with current and future LIBs. The model facilitates the assessment of various technologies and technology chains with regards to manufacturing cost. Hence, critical process steps in the production of ASSBs could be identified and further development needs were derived.

### Formal Requirements

In addition to the specific requirements, also the fulfillment of the formal requirements will be critically reflected in the following:

- *Reasonable benefit-to-cost ratio:* As outlined in subsection 5.2.1, the benefits of applying the methodology can outweigh the associated cost by several orders of magnitude.
- *Modularity of the methodology:* The modular structure of the methodology allows to further reduce the effort by adjusting the level of detail for the individual modules. In the application example, the analysis of the company's technology strategy revealed a relatively high risk affinity. Hence, in contrast to the detailed assessment of the technology maturity suggested by REINHART & SCHINDLER (2010), a rough estimation of the technology maturity seemed sufficient for technology preselection within the AHP. Furthermore, by providing an overview of potentially suitable technologies for ASSB production (cf. **P 2** and **P 3**), the effort for technology identification can be significantly reduced and the modular structure allows for a simple integration of new technologies into the technology pool.
- *Transparency of the methodology:* The provided methods for technology evaluation and preselection allow for a transparent decision making process. The matrix-based approach using the AHP facilitates a profound

selection of suitable technologies based on a multi-criteria assessment (cf. P 3). In particular the bottom-up calculation model developed in P 4 allows to trace back the cost to each individual process step along the technology chain. Hence, critical process steps can be identified and targets for further development can be quantified.

- *Reusability of the methodology:* Within the application example, the methodology was adapted towards the specific requirements of the battery cell manufacturer. Thus, different materials and new technologies could be integrated and evaluated. The modular structure and in particular the design review stage also allow for an iterative procedure to successively refine the product design and the corresponding technology chains. Hence, emerging technologies can be easily integrated and evaluated to update the relevant information with an increasing knowledge base. Of course, all relevant interactions need to be taken into account during technology chain generation, e.g. by updating the suggested heuristic procedure (cf. subsection 4.1.3) or using matrix-based methods.
- *Generality of the methodology:* The methodology has been applied to a multitude of processing routes for various types of ASSBs with different inorganic SE materials. Furthermore, within the application example, the methodology was easily adapted towards polymer-based SEs. This confirms the required generality of the methodology. Due to the focus on existing cell formats as defined by DIN 91252, a certain bias towards conventional LICs cannot be excluded. As shown in literature, ASSBs could also allow to fabricate ultra-thick electrodes—up to 600  $\mu\text{m}$  (KATO ET AL. 2018)—and three-dimensional designs as for microbatteries (OUDENHOVEN ET AL. 2011). This would allow for completely different cell concepts which have not been considered within this thesis.

While the methodology could be successfully verified within the application example, the validity of the results yet needs to be confirmed. Since all input for the hypothetical scenarios was based on literature and expert knowledge, pilot plant equipment will be required for an experimental validation. The results of this thesis can help to focus research efforts and spend resources based on substantiated decisions. Overall, the specific and formal requirements on the

methodology for strategic planning of technology chains for ASSB production have been met to a large extent.

### 5.2.3 Critical Discussion of Research Questions and Hypotheses

Within chapter 2, hypotheses were defined in order to support the overall aim of this thesis, i.e. to generate knowledge at the interface between laboratory research and industrial mass production. Based on the results of the publications **P 1** to **P 4** and the application example, these hypotheses will now be discussed and confirmed or rejected if possible, which also serves to shed light on the research questions defined within section 1.3:

- **H 1** *For oxide-based ASSBs, sintering is a critical process step from a technical and economic perspective:* The sintering step for oxide-based ASSB is indeed critical. On the one hand, undesired side reactions of cathode and SE necessitate a complicated cell design (cf. **P 2**) which brings along further challenges for post-processing (cf. **P 3**). Furthermore the shrinkage during sintering and the fragility of the resulting sintered sheets can lead to high scrap rates which significantly contribute to the overall manufacturing cost (cf. **P 4**). Also the sintering process itself results in high cost due to the large plant area required for the furnaces and the high energy consumption (cf. TROY ET AL. (2016) and **P 4**). Hence, the hypothesis can be confirmed.
- **H 2** *For sulfide-based ASSBs, the processing atmosphere is a critical influence factor from a technical and economic perspective:* The risk of generating toxic H<sub>2</sub>S during production or use of sulfide-based ASSBs must be reduced to a minimum. Processing under inert gas results in complicated intralogistics, in particular with regards to the work piece carriers and vacuum locks. This also impedes cleaning of the equipment and troubleshooting in case of break-downs. Furthermore, a proper functionality of electric drives in Argon atmosphere is questionable and needs to be individually tested for each device integrated into the glovebox. Nonetheless, in contrast to common understanding, the calculations in **P 4** reveal that the overall cost (investment, operating supplies, etc.) is not significantly higher than processing in a dry room. Recent research also indicates that H<sub>2</sub>S generation can be significantly reduced by a combination of sulfide-based

SE with a suitable polymer, potentially also allowing for processing in a dry room (TAN ET AL. 2019). Further empirical research will be required to conclusively confirm or refute the hypothesis.

- **H 3** *Vapor deposition or spray coating techniques are promising technologies for SES fabrication:* As deduced in **P 3**, most vapor deposition and spray coating technologies are not suitable to fabricate SES layers in the relevant geometries due to high error rates or processing temperatures. While the ADM seems promising from a technical perspective to fulfill product requirements and circumvent the critical sintering step for oxide-based SEs, the technology maturity is currently too low for an industrial application. Hence, a lot of development effort will be necessary to increase deposition efficiencies and enable the throughputs required for an economic fabrication (cf. **P 4**). Therefore, the hypothesis needs to be rejected unless a major breakthrough in technology development is realized.
- **H 4** *For oxide-based ASSBs, infiltration of cathode materials into a porous, sintered SE host is a promising approach:* While the issues associated with co-sintering of cathode materials with oxide-based SE can to a certain extent be circumvented by infiltration of cathode materials after fabricating a sintered host structure, several challenges have still not been satisfactorily addressed. From a technical perspective, careful tuning of porosities and infiltration parameters is necessary to achieve dense layers with sufficiently high cathode loading. Hence, multiple infiltration and calcination steps may be required (REN ET AL. 2017). Furthermore, remaining issues at the interface between cathode and SE particles remain unresolved. While the manufacturing cost could be pushed towards 150 \$/kWh by economies of scale (cf. **P 3**), competitiveness with conventional LICs or sulfide-based ASSBs can hardly be reached on cell level (**P 4**). Hence, unless significant advantages on the battery module or pack level can be achieved, the hypothesis must be rejected.
- **H 5** *Manufacturing and processing of thin lithium metal films are critical both technically and economically:* Processing of lithium metal is critical from a technical perspective—safety issues occur due to the high reactivity, and the adhesiveness complicates handling and thinning down towards relevant thicknesses. A homogeneous deposition for melt- or vapor-based processing and electrochemical precipitation is strongly dependent on the

substrate surface. Despite the uncertainty about material cost, the higher energy density for battery cells with lithium anode can result in lower cost than for conventional graphite or Si/C anodes (cf. P 4). Thus, the hypothesis can only partly be confirmed.

- **H 6** *Material supply and cost are critical for ASSB production:* Currently, only few material suppliers actually offer SE materials on the market, and only small quantities (a few g to kg) are sold with fluctuating quality. As shown both within the top-down calculation in P 3 and the bottom-up calculation in P 4, the SE material cost has a strong impact on the overall ASSB battery cost, which is in good agreement with estimations by SCHMUCH ET AL. (2018). In general, the cost impact is higher for SE materials with a higher density (cf. P 4). The hypothesis can therefore be confirmed.

To conclude, the results of this thesis help to clarify many of the assumptions frequently made in the literature on ASSB production and cost (cf. chapter 2). By providing a suitable methodological framework and shedding light on the above described hypotheses, answers can be provided to the research questions defined in subsection 1.3.1: Based on the performance measures and material properties outlined in section 2.1 and section 2.2, requirements for industrial fabrication of ASSBs (Q 1) have successfully been gathered within P 2: These include, amongst others, physical and chemical properties (such as ionic conductivity or reactivity), geometrical considerations (such as layer thicknesses and compositions), mechanical properties (such as bending stiffness), production environment (such as inert gas), material cost, etc. Furthermore, technologies and technology chains for ASSB production (Q 2) were systematically assessed based on technical and economic criteria (cf. P 3 and P 4), with a particular focus on manufacturing of the different ASSB components: For the SES and cathode, wet chemical processing seems to be most likely for both sulfide- and oxide-based ASSBs. However, technologies enabling a solvent reduction could allow for a cost savings potential. Similarly, a lithium metal anode could provide an economic benefit, although further research will be required to conclude on the manufacturing technologies best suited. Finally, critical factors in the production of ASSBs (Q 3) could be identified with the aid of the above discussed hypotheses. This is a prerequisite to mitigate the risks and reduce the complexity associated with the high uncertainty on industrial production of large-format ASSBs. The methodology developed within this thesis can help

## 5 Discussion—Application and Evaluation

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to successively refine the results with ongoing development and a broader knowledge base on ASSBs.

### 5.3 Summary

In conclusion, the applicability of the methodology was successfully verified in collaboration with an international battery cell producer. The application example confirms that the specific and formal requirements deduced in section 3.1 have been met to a large extent. Finally, the discussion of the hypotheses defined in chapter 2 significantly contributes to the knowledge base on ASSB production and therefore helps academic and industrial stakeholders involved in the scale-up towards pilot production. The following chapter will conclude with a summary of the thesis and an outlook on future research.

## **6 Conclusion**

Efficient and affordable means of energy storage are required to enable the transfer from fossil fuels to renewable energies. Despite the great potential for safer batteries with better performance measures, not much has been known on the challenges and possible solutions for industrial production of large-format all-solid-state batteries (ASSBs). Hence, this thesis has made a significant contribution in the research field by providing guidelines for stakeholders in the upscaling from laboratory to pilot and industrial scale. The main results of this thesis will be summarized in the following section, followed by an outlook on possible future research.

### **6.1 Summary of the Thesis**

Since current know-how on ASSBs mainly comes from laboratory research, the objective of this thesis is to generate knowledge at the interface between fundamental sciences and production engineering, and to assist stakeholders involved in the scale-up of production processes for the fabrication of large-format ASSBs. Therefore, within the defined framework, this thesis served to provide answers to the three research questions: What are requirements for industrial fabrication of ASSBs? Which technologies and technology chains are suitable for ASSB production? Which factors are critical in the production of ASSBs? Due to the applied research focus at the interface between laboratory research and strategic technology planning, the Design Research Methodology (DRM) by BLESSING & CHAKRABARTI (2009) served as a guidance throughout this work.

The literature review on the current state of research on ASSBs revealed a high uncertainty associated with the multitude of solid electrolyte (SE) materials and an ambiguous cell design, challenges at the interfaces between different materials, and a large variety of technologies employed on the laboratory scale to manufacture ASSB components. Moreover, only little know-how exists from a production engineering perspective, and investments in the scale-up of ASSB production are associated with a high risk due to the expected competition with the conventional lithium-ion battery (LIB) technology. Hence, a systematic and

## 6 Conclusion

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structured procedure is required to tackle the current challenges for ASSB scale-up and to confirm or refute the hypotheses formulated to refine the underlying research questions.

Therefore, requirements on a methodological approach for strategic technology planning for ASSB production were defined. A critical analysis of the state of the art in strategic planning of technology chains and battery production cost modeling revealed that existing methods must be adapted towards the specific requirements for ASSB production.

Hence, a methodology for strategic planning of technology chains for the production of large-format ASSBs was developed. The four peer-reviewed publications (P 1 to P 4) summarized within this thesis served to highlight the main models and methods, which were also used to derive answers to the research questions and hypotheses defined beforehand: A product model was developed in P 1 to allow for a structured requirements analysis. This serves as an input for the identification and evaluation of production technologies. Taking into account the relevant interactions, technology chains for ASSB production are generated and evaluated. Based on an expert elicitation, different cell designs and processing routes for ASSB were gathered and summarized in a comprehensive study (P 2). This served, amongst others, to compare an exemplary technology chain for a sulfide-based ASSB with conventional lithium-ion cell (LIC) production: Major differences are the fabrication of the solid electrolyte separator (SES) and the lithium metal anode, as well as the necessity to produce in an inert gas atmosphere (glovebox environment). In P 3, various manufacturing technologies for ceramic based energy systems were analyzed with regards to their applicability for oxide-based ASSBs: While established wet chemical technologies (e.g. tape casting or screen printing) seem to be advantageous with regards to production related aspects, the need for a high temperature sintering requires elaborate cell design. The aerosol deposition method (ADM) seems a promising alternative since it could render the high temperature sintering step void. However, the technology maturity is still far from an industrial application. A top-down calculation was undertaken, revealing that SE material cost and elaborate cathode design are critical for oxide-based ASSBs to become competitive with conventional LICs. In order to enable a detailed economic comparison with conventional LIC production and

to identify critical process steps, a bottom-up cost modeling tool was developed in P 4. The calculations reveal that sulfide-based ASSBs can indeed become competitive with conventional LIC if material issues are resolved and production is successfully scaled. In contrast, an economic fabrication of oxide-based ASSBs will be challenging due to the high material and sintering cost.

Finally, the discussion of the results helped to verify the applicability of the methodology and to critically reflect the outcomes of this thesis with regards to the requirements derived from the current state of research on ASSBs. The developed methodology was successfully transferred to an industrial application with an established battery cell manufacturer. The evaluation revealed that the methodology can provide an economic benefit and the specific and formal requirements are mostly or completely fulfilled. Furthermore, by shedding light on the earlier defined hypotheses, answers could be provided to the underlying research questions. Although a validation of the results would require further investigations using pilot scale equipment, a sound basis was created to efficiently tackle the next steps towards the production of large-format ASSBs.

To conclude, in order for ASSBs to succeed on the market, the energy density, fast-charging behavior, cycle life, safety, and cost must become competitive to conventional LIBs. Selecting the right production technology (chain) not only significantly impacts the manufacturing cost, but also the other performance measures by setting constraints for the ASSB cell design. Thus, by providing a suitable methodological framework, this thesis can help stakeholders in research and industry to make profound decisions and focus their development efforts on the most promising scenarios. Furthermore, the author's work significantly adds to the state of research on ASSBs by extending the knowledge base from a production engineering perspective and calculating the expected ASSB manufacturing cost using both a top-down and a bottom-up approach. Hence, this thesis can considerably contribute to the development of better batteries for improved energy storage.

## 6.2 Future Research

The results of this thesis represent a major step to pave the way to ASSB mass production. By bringing together product and process development within the framework of Simultaneous Engineering, the foundation was laid for further

## 6 Conclusion

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research to validate the findings of this work. This comprises in particular experimental investigations on the fabrication, processing, and handling of ASSB components. Of course, further steps towards industrial production will need to be accompanied by an implementation of scalable production processes on the pilot scale. While the focus of this thesis is mainly on different scenarios for ASSB components manufacturing, the results can easily be extended to ASSB cell assembly (KONWITSCHNY ET AL. 2019). This will be particularly important for automated handling of lithium foils and brittle composite electrode and SES sheets or multi-layer systems. Attention should also be paid to the assembly back-end, i.e. (automated) contacting, packaging, and formation of large-format ASSBs. This will also facilitate the integration into ASSB modules and packs.

The findings of the author's work have already been successfully transferred into practical application: First experimental results have been obtained, for instance, by investigating the processability of sulfide-based SES layers (RIPHAUS ET AL. 2018). The work confirms that roll-to-roll processing using established wet coating and calendaring technologies is possible by employing suitable binder systems. Furthermore, based on an analysis of different processing routes for ASSB cell assembly, a handling test rig for ASSB components was developed and built up at the *iwb* (KONWITSCHNY ET AL. 2019). Further research will take place within the research project *Produktionstechnik für Festkörperzellen mit Lithium-Metall-Anode* (Engl. *Production technology for all-solid-state batteries with lithium metal anode*), funded by the Federal Ministry of Education and Research (Bundesministerium für Bildung und Forschung), grant number 03XP0184I (ProFeLi). This creates a sound basis for a continuation of the work.

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Zhu, Y.; He, X.; Mo, Y.: First principles study on electrochemical and chemical stability of solid electrolyte–electrode interfaces in all-solid-state Li-ion batteries. *Journal of Materials Chemistry A* 4 (2016) 9, pp. 3253–3266.

## Appendix

## A.1 Individual Contributions to the Embedded Publications

The author’s individual contribution to the embedded publications are summarized in table A.1. All the publications have been developed in close collaboration with the co-authors, whose contribution is highly appreciated.

*Table A.1: Individual contributions of the author to the publications P 1–P 4.*

	<b>P 1</b>	<b>P 2</b>	<b>P 3</b>	<b>P 4</b>
Planning and coordination	90 %	70 %	95 %	95 %
Literature review and analysis	90 %	95 %	70 %	95 %
Method development	60 %	60 %	70 %	70 %
Data collection	n/a	n/a	70 %	80 %
Expert elicitation	60 %	70 %	60 %	70 %
Modeling and calculation	50 %	n/a	70 %	80 %
Interpretation of results	70 %	90 %	90 %	90 %
Preparation of figures and tables	70 %	95 %	70 %	95 %
Writing of the publication	80 %	90 %	80 %	95 %

## A.2 Theses Supervised

In the context of the research work performed by the author—which is described both in the embedded research publications P 1 to P 4 and this dissertation—various Bachelor’s theses, semester projects, and Master’s theses have been intensively supervised methodically and with regard to the development of their research clarification, problem statements, objectives, research questions, and content. The supervision took place at the *iwb* of the Technical University of Munich (TUM). Bachelor’s and Master’s theses as well as seminar papers listed in table A.2 contributed to the author’s dissertation (in chronological order). As the student’s theses have been elaborated in close relationship with the author’s research activities, their topics are all located within the field of strategic technology planning for ASSB production. Findings and results of these collaborative research projects have partly contributed to this dissertation and the publications P 1 to P 4. The author would like to express his sincere gratitude for the remarkable commitment of all his students and their valuable and interesting contributions.

## A.2 Theses Supervised

Table A.2: Theses supervised within the course of this research project.

Name	Title	Period	Contribution
Christoph Vieider	Konzept zur Industrialisierung von Festkörperbatterien für die Elektromobilität	04/2016-10/2016	<b>P 2</b>
Alexander Just	Fertigungs- und montagegerechtes Design für Festkörperbatteriezellen	10/2016-04/2017	<b>P 2</b>
Larissa Köhler	Systematischer Vergleich der Prozessketten zur Herstellung von Festelektrolytzellen mit der konventionellen Batterieproduktion	10/2016-05/2017	<b>P 2</b>
Célestine Singer	Analyse der Herstellungsverfahren oxidkeramischer Energiesysteme bezüglich ihrer Anwendbarkeit in der Produktion von Festkörperbatterien	11/2016-05/2017	<b>P 1, P 3</b>
Joseph Nefzger	Identifikation von Produktionstechnologien für Festkörperbatterien auf Basis eines Produktmodells	04/2017-09/2017	subsection 4.1.1, subsection 4.1.2, subsection 5.1.2
Anselm Mülberger	Methodik zur strategischen Planung von Prozessfolgen für die Herstellung von Festkörperbatterien	10/2017-03/2018	section 3.3, section 4.1, subsection 5.1.2
Kathrin Mosler	Verarbeitung von Materialien zur Herstellung von Festkörperbatterien für die Elektromobilität	10/2017-04/2018	section 6.2
Heiko Knörzer	Entwicklung eines Kostenmodells für die industrielle Herstellung von Festelektrolytzellen	10/2017-05/2018	subsection 3.3.1, subsection 4.1.5, <b>P 4</b>
Franziska Peteler	Fertigungstechnologien für Festkörperbatterien: Technische Eignung und wirtschaftliche Priorisierung	12/2017-07/2018	subsection 5.1.2
Julia Aulbach	Methode zur Bewertung des Technologiepotenzials am Beispiel alternativer Prozessketten für die Herstellung von Feststoffbatterien	05/2018-08/2018	subsection 4.1.2
Célestine Singer	Entwicklung eines Systemmodells für die strategische Planung von Technologieketten zur Herstellung von Festkörperbatterien	06/2018-12/2018	section 2.2, section 2.3, section 3.1, subsection 3.3.1, section 4.1, subsection 5.1.2, <b>P 3</b>
Heiko Knörzer	Entwicklung eines flexiblen Kostenmodells für den Vergleich von industriellen Herstellungsverfahren von Festelektrolytzellen	07/2018-01/2019	subsection 3.3.1, subsection 4.1.5, subsection 5.1.2, <b>P 4</b>
Anna Julia Imbsweiler	Industrialization Study for the Economic Fabrication of All-Solid-State Batteries	10/2018-04/2019	subsection 5.1.2, <b>P 4</b>

### A.3 Publication List

During his work at the *iwb*, the author contributed to the following publications:

GÜNTHER ET AL. (2016)

Günther, T.; Billot, N.; Schuster, J.; **Schnell, J.**; Spingler, F. B.; Gasteiger, H. A.: The Manufacturing of Electrodes: Key Process for the Future Success of Lithium-Ion Batteries. *Advanced Materials Research* 1140 (2016), pp. 304–311.

KNOCHE ET AL. (2016)

Knoche, T.; Zinth, V.; Schulz, M.; **Schnell, J.**; Gilles, R.; Reinhart, G.: In situ visualization of the electrolyte solvent filling process by neutron radiography. *Journal of Power Sources* 331 (2016), pp. 267–276.

SCHNELL & REINHART (2016)

**Schnell, J.**; Reinhart, G.: Quality Management for Battery Production: A Quality Gate Concept. *Procedia CIRP* 57 (2016), pp. 568-573.

REINHART ET AL. (2017)

Reinhart, .; Knoll, D.; Teschemacher, U.; Lux, G.; **Schnell, J.**; Endres, F.; Distel, F.; Seidel, C.; Berger, C.; Klöber-Koch, J.; Pielmeier, J.; Braunreuther, S.: Anwendungsfeld Automobilindustrie. In: Reinhart, G. (ed.): *Handbuch Industrie 4.0. Geschäftsmodelle, Prozesse, Technik*. München: Hanser, 2017, pp. 709–722, ISBN: 978-3-446-44989-3.

SCHNELL ET AL. (2017)

**Schnell, J.**; Hofer, A.; Singer, C.; Günther, T.; Reinhart, G.: Evaluation of technology chains for the production of all-solid-state batteries. In: Schmitt, R.; Schuh, G. (eds): *7. WGP-Jahreskongress Aachen*, October 5-6, 2017. Aachen, Apprimus Verlag, 2017, pp. 295-302, ISBN: 978-3-86359-555-5.

RIPHAUS ET AL. (2018)

Ripphaus, N.; Strobl, P.; Stiaszny, B.; Zinkevich, T.; Yavuz, M.; **Schnell, J.**; In-dris, S.; Gasteiger, H. A.; Sedlmaier, S. J.: Slurry-Based Processing of Solid Electrolytes: A Comparative Binder Study. *Journal of the Electrochemical Society* 165 (2018) 16, pp. A3993–A3999.

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KONWITSCHNY ET AL. (2019)

Konwitschny, F.; **Schnell, J.**; Reinhart, G.: Handling Cell Components in the Production of Multi-Layered Large Format All-Solid-State Batteries with Lithium Anode. *Procedia CIRP* 81 (2019), pp. 1236-1241.

SCHNELL ET AL. (2019a)

**Schnell, J.**; Tietz, F.; Singer, C.; Hofer, A.; Billot, N.; Reinhart, G.: Prospects of production technologies and manufacturing costs of oxide-based all-solid-state lithium batteries. *Energy & Environmental Science* 12 (2019) 6, pp. 1818–1833.

SCHNELL ET AL. (2019b)

**Schnell, J.**; Nentwich, C.; Endres, F.; Kollenda, A.; Distel, F.; Knoche, T.; Reinhart, G.: Data mining in lithium-ion battery cell production, *Journal of Power Sources* 413 (2019), pp. 360–366.

SCHNELL ET AL. (2020)

**Schnell, J.**; Knörzer, H.; Imbsweiler, A. J.; Reinhart, G.: Solid versus Liquid—A Bottom-Up Calculation Model to Analyze the Manufacturing Cost of Future High-Energy Batteries. *Energy Technology* (2020) p. 1901237.



## **Eidesstattliche Erklärung**

Ich erkläre hiermit eidesstattlich, dass ich die vorliegende Arbeit selbstständig angefertigt habe. Die aus fremden Quellen direkt oder indirekt übernommenen Gedanken sind als solche gekennzeichnet.

Die Arbeit wurde bisher keiner anderen Prüfungsbehörde vorgelegt.

Garching, den 18.06.2020

(Joscha Schnell)