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Handling Cell Components in the Production of Multi-Layered Large Format All-Solid-State Batteries with Lithium Anode

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Abstract

All-solid-state batteries (ASSB) are considered as promising energy storage systems for consumer electronics and electric mobility because of their high safety, long life and high energy density. Currently, electrochemistry and materials research attempt to produce ASSB cells on laboratory scale. One future challenge will be to characterize the required manufacturing processes for large format composites of thin ceramic layers used in ASSB and to transfer this knowledge to pilot scale production. The promising technology is still far from being commercially producible on an industrial scale, which is why there is a particular need for extensive research on the manufacturability of large format and stackable sheet type ASSB cells. In this paper we present a concept for handling cell components during cell assembly of ASSB by analyzing extensively the material and process requirements, as well as interdependencies with simultaneous engineering. Furthermore, the possibilities for cell stacking are investigated via SIPOC by appropriately adapting techniques of conventional lithium-ion batteries to the varying requirements of ASSB.

Keywords: all-solid-state battery; ASSB; battery production; solid electrolyte; concept; handling system; gripping system; lithium metal anode; DFA; SIPOC

1. Introduction

Lithium-ion batteries are a key enabler for emission free mobility. However, the ever increasing demand for higher energy densities has pushed the current technology towards its limit [1]. The implementation of all-solid-state batteries (ASSB) could facilitate the use of a lithium (Li) metal anode, potentially enabling much higher energy densities than the current technology [2]. Furthermore, the omission of flammable liquid components from the battery cells promises less effort for safety management: Typically, the electrolyte liquid soaks the porous media in a lithium-ion cell to enable ion transport from the cathode via the separator to the anode and vice versa. Replacing the separator (and also the electrolyte in the electrodes) with a non-flammable solid electrolyte could, therefore, circumvent the risks of leakage and flammability. Some solid electrolytes show relatively high ionic conductivities, some of them even exceeding those of conventional liquid electrolytes [3].

Nomenclature

ASSB	All-solid-state battery
LIB	Lithium-ion battery/ Lithium-ion battery cell(s)
SE	Solid electrolyte
CC	Cathode composite
Li	Lithium
SIPOC	Supplier input process output customer analysis
DFM/A	Design for manufacturing/ assembly
PLC	Programmable logic controller

Despite the great potential, there is still an ongoing need for research on ASSB, especially with regard to the unresolved

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issues at the interfaces between electrodes and solid electrolyte [4].While a lot of effort has been put into tackling the issues on material level, reports on the scale-up from laboratory to pilot and industrial scale remain scarce [5,6]. An increasing number of publications have been dealing with scalable processes for the fabrication of composite cathode layers with high active material loading [7,8] and thin (< 30 µm) solid electrolyte layers [9,10]. Currently, these components are assembled manually to fabricate ASSB cells on laboratory scale [10,11]. While fully automated winding and stacking processes have been widely established for conventional lithium-ion cells [12,13], challenges are expected for automated assembly of large format ASSB cells: The mechanical properties of ASSB components, i.e. the brittleness and low bending stiffness of composite cathodes and solid electrolyte layers with low porosities [1] may not allow for conventional winding or folding processes. Furthermore, the sensitivity and adhesiveness of lithium metal as anode material can turn out to be challenging for conventional handling technologies such as vacuum-based suction grippers. Hence, investigations on automated handling of ASSB components will be required to enable simultaneous engineering of product design and production processes for large format ASSB.

The scope of this paper is, therefore, the analysis of challenges and requirements for an automated assembly of ASSB cells. First, an explanation of the materials used and associated cell components is given, as well as a description of possible cell design variants. Based on this, the differences and challenges for the production of ASSB compared to conventional lithium-ion batteries (LIB) are analysed. Subsequently, process and system requirements for handling and assembly procedures are derived from the properties of the specific components used in ASSB cells. In the last section, a concept for the development of a handling system for ASSB components during cell assembly is described and the implementation of a handling test rig for carrying out handling tests with ASSB components is suggested.

2. Fundamentals: components, stacking and cell design

2.1. Solid electrolyte and separator

Solid electrolytes (SE) combine two functions: firstly, the ionic conductivity, which allows the transportation of Li ions from anode to cathode, and secondly the electric isolation between these two [14]. The solid electrolyte hence substitutes the liquid electrolyte and the separator when comparing it to a conventional LIB. Solid electrolyte systems can be subdivided into polymeric and inorganic electrolytes [14]. Due to their promising properties with respect to ASSB, the focus of this paper will be on sulfide and oxide based solid electrolytes.

2.2. Anode

With regard to anode materials for ASSB, lithium (Li) metal is considered as one of the most promising candidates [14] because it could lead to a high energy density of the cell. However, the coulombic efficiency during deposition and retrieval of lithium is limited [15]. The Li layer needs to be very thin (~10 µm) in order to achieve high energy densities. The formation of Li dendrites, caused by undefined deposition of Li during battery discharge, remains an issue. Remedial action can be taken by applying a protective layer [17].

2.3. Cathode

A composite cathode (CC), consisting of cathode active material, solid electrolyte, conducting agent will be required in order to allow for sufficient ionic and electric percolation. Furthermore, suitable binders will be required to ensure mechanical stability and long lifetime of the ASSB [4]. A lack of mechanical contact between the particles of SE, CC and anode leads to high transition resistances between the SE layer and the electrodes and poor cycle stability.

To achieve low resistances, better ionic conductivity and a competitive energy density in contrast to LIB, a high cathode active material content and thin SE layers are required.

2.4. Design possibilities for the superstructure of ASSB cells

One-layer systems with only one galvanic cell are mainly used for thin film batteries, where only low energy density is required. In contrast, multi-layer systems consist of multiple galvanic cells which are stacked and electrically contacted with each other. Only a parallel interconnection of galvanic cells is possible for conventional LIB due to the homogenous distribution of the liquid electrolyte. In contrast, the following two design possibilities (Fig. 1) emerge for ASSB:

1. Parallel cell stack:

In a parallel stacking configuration, the single galvanic cells are stacked in such way that always the corresponding electrical poles of two galvanic cells face each other. They are connected to each other by using the same current collector tab for supplying the current to and draining it from the galvanic cell(s). [18,19] This stacking concept functions according to the same principle as conventional LIB with conventional "monopolar" current collectors. Nonetheless, it must be further examined whether the stacking process for parallel stacked ASSB is also comparable to the one of conventional LIB.

2. Bipolar cell stack:

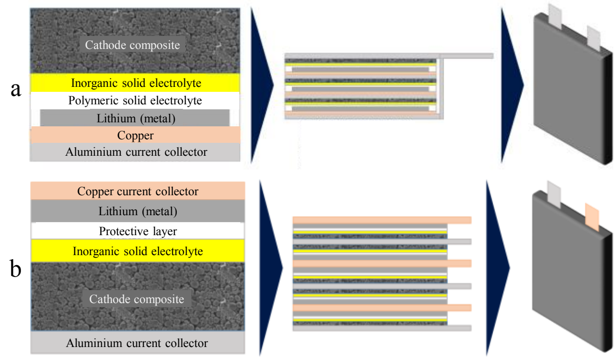

Fig. 1. (a) bipolar [21]; (b) parallel stacking of galvanic ASSB cells.

In a bipolar stacking configuration, the single galvanic cells, consisting of active electrode materials, solid electrolyte layers and current collectors, are stacked and simultaneously connected in series to each other [18,20,21].

This promising cell stacking method enables the generation of high voltage cells but requires bipolar current collectors in return. These must somehow ionically isolate the two adjoining electrodes and are electrochemically stable to electrode materials at their specific potentials.

3. Derivation of process and system requirements

The process requirements regarding the handling of the abovementioned components in ASSB can be defined more systematically by considering the following aspects.

3.1. Aspects regarding production and assembly technology

A major goal within battery cell production is to keep the cell assembly processes as simple as possible in order to ensure high quality of the cell components. Additionally, the processes need to be as fast possible in order to maximize throughput and, thus, minimize production costs. These objectives also apply to the production of ASSB. Approaches and possibilities to achieve these goals are: reducing non-value-adding handling operations, transport processes and storage phases during cell assembly [22]; simplifying the product and the joining of components [22]; designing the product assembly-compliant or assembly-oriented in accordance with the guidelines regarding a design for manufacturing and assembly (DFM/DFA) [23].

In particular, DFA methods need to be applied very early in the product development process (already during the phase of product engineering and product development) since product changes at this stage can have a major influence on the manufacturing and assembly processes [22]. In order to link the product development of novel ASSB with the development of the corresponding production processes, the application of DFA guidelines is highly relevant, already during the specification of stack and cell design.

3.2. (Electro)chemical aspects

Under the premise of influencing the electrochemical performance of the cell and its components as little as possible, it is of great importance to understand which material modifications can occur during the handling and stacking process. The chemical reactivity of the processed materials influences the handling technology and beyond that the applied gripping principle. For example, if the cell components are chemically unstable towards the materials used for the gripper it may be necessary to change the material of the gripper to prevent negative effects on the cell components.

Another aspect in this context are reactions of the processed materials with the surrounding atmosphere during handling and stacking. Any contact with moisture (e.g. in the ambient) air before, during and after these processes can lead to a chemical reaction and thus a degradation of the material [1]. Pure Li [24] and sulfidic SE [25] systems are particularly susceptible to this, but oxidic systems [26] may also react sensitively.

3.3. Safety aspects

Sensitivity to moisture is also important from a safety point of view when handling ASSB components. The entire process chain of cell assembly must therefore be executed in a specially dehumified atmosphere (dry room) or even under inert gas atmosphere (e.g. glove box flooded with argon) to ensure the required safety for processing critical materials and also to prevent negative effects on an electrochemical level (see 3.2). The development and design of technical protective measures for both the processed materials as well as the production facilities is subject to consideration during the design of a handling and stacking system, especially for large format ASSB.

3.4. Mechanical aspects

The different cell components of ASSB differ significantly with regard to their mechanical and physical properties. Due to the targeted low porosities, the CC and SE layers are most likely sensitive to mechanical contact or pressure, shock, vibration and bending. For this reason, particular care must be taken to ensure gentle handling of these components. The integration of adequate binders could provide more flexibility [27].

In terms of mechanical properties, lithium is very adhesive and therefore easily sticks to a gripper or any mechanical guidance during processing. Another challenge will be the required thickness of the Li film which needs to be only a few micrometers. Thereby the film is easily deformable and mechanically unstable, the feasibility of a self-supporting Li foil becomes difficult. Rather, pre-"lithiated" carrier layers (either a current collector or the SE) to which the lithium applies by lamination or evaporation will be the object of handling operations.

3.5. Quality aspects

The quality of the final ASSB cell strongly depends on the electrochemical functionality of its components in the single galvanic cells and the entire cell stack. A gentle handling of the components is required to minimize negative effects on the electrochemical functionality. The lowest possible mechanical impact on the component should be achieved at this. Therefore, a quality-related design of the components handling and the corresponding handling system must be carried out.

The other important aspect for ensuring good cell quality is the deposition accuracy of the automatic handling machine during stack formation. The more accurately the components can be stacked, the better the electrochemical performance of the galvanic cell but also of the entire ASSB cell.

3.6. Economical aspects

The key target for handling and processing of ASSB components is the greatest possible yield and throughput, an economic evaluation is therefore subject of further research.

4. Production of ASSB: challenges and changes compared to conventional LIB

In contrast to conventional assembly planning within LIB production, the product design for ASSB can currently only be vaguely described. Based on the method for evaluation of technologies for ASSB production [28], different approaches for the fabrication of ASSB layers were summarized by Schnell et al. [6]. However, the assembly processes are only roughly outlined and therefore require further investigation. In order to categorize the different possibilities for ASSB stack assembly, a morphological box was created, as illustrated in Table 1. On the basis of Reinhart et al. [29] the structure follows a SIPOC (supplier-input-process-output-customer) approach:

Table 1: Morphological box following SIPOC.

SIPOC	Cluster	Possibilities			
Supplier	Components	Layer fabrication processes	External supplier		
Input	Type	Sheets	Rolling goods		
	Combination	Separate components	Half cells	Galvanic cells	
	Solid electrolyte separator	On carrier foil	Self-supporting	On cathode	
	Anode	On carrier foil	Self-supporting	On current collector	On solid electrolyte
	Cathode	On carrier foil	Self-supporting	On current collector	
	Current collector	Self-supporting	On anode		
	Optional: Protective layer	On solid electrolyte separator	On anode		
Process	Integration	Combined with sheet cutting	Separate processes		
	Principle	Sheet stacking	Z-folding	Flat winding?	
	Atmosphere	Inert gas	Dry room		
	Positioning	In work piece carrier	Into pouch foil	Into hard case	
	Gripper	Vacuum	Contactless	others	
Output	Stack design	Bipolar	Parallel		
	Number of layers	Variable			
	Geometry	Variable			
Customer	Assembly backend	Welding/ contacting	Stack pressing	Packaging	

The suppliers of the stacking process are all preceding processes (i.e. layer fabrication) and external suppliers for component manufacturing. The components can either be provided as single- or multi-layer sheets (e.g. after sintering) or as rolling goods (e.g. after calendaring). The amount of handling steps per "full cell" can be drastically reduced if the components are already supplied as "half cells" or even galvanic cells, i.e. the cathode, solid electrolyte separator, and anode laminated on top of each other. However, the latter involves the risk of short circuiting during cutting and assembly since the cells will already be in a charged state if lithium is employed as anode material. Depending on the mechanical properties of the different components and the preceding manufacturing processes, the components can either be provided as free-standing layers, on a carrier foil (to be peeled off), or combined with one or several of the neighboring components. For instance, for hybrid solid electrolytes with sufficient mechanical flexibility, self-standing layers or a carrier foil as substrate are conceivable [27], while otherwise the solid electrolyte layer needs to be directly coated onto one of the electrodes (i.e. the cathode, taking into account that lithium foil will probably not be suitable as substrate) [30].

The material supply also has consequences for the layout of the stacking process: While a flexible hybrid solid electrolyte separator could possibly allow for z-folding or even flat winding, a single sheet stacking process will be required for rigid components with low bending stiffness. A special focus needs to be drawn to the surfaces to be handled: For instance, a damage-free manipulation of the solid electrolyte separator is vital for proper functionality and safety of the cell. An integration of the stacking process with the preceding cutting step can reduce the effort for intermediate storage and multiple handling steps. Depending on the materials used, a dry room or even inert gas atmosphere will be required. This can also have an impact on the gripping principle, e.g. additional effort for inert gas recycling when employing vacuum-based suction grippers. Positioning of the layers directly into the housing can help to reduce further handling steps during assembly backend.

The final ASSB stack with its defined geometry and number of layers is then provided to the customers of the process, i.e. the following processes in the assembly backend (e.g. tab welding and sealing).

5. Development of a handling system for cell assembly

5.1. Conceptual design

A multi-disciplinary planning approach is proposed to encompass the different aspects influencing the fabrication of multi-layered ASSB, as well as to incorporate them into the ASSB design and the development of appropriate handling procedures and machinery for cell stacking and assembly. As shown in Fig. 2, the framework consists of three major aspects: "new product development", "process engineering" and "systems engineering", which must be closely interlinked in order to master the aforementioned challenges.

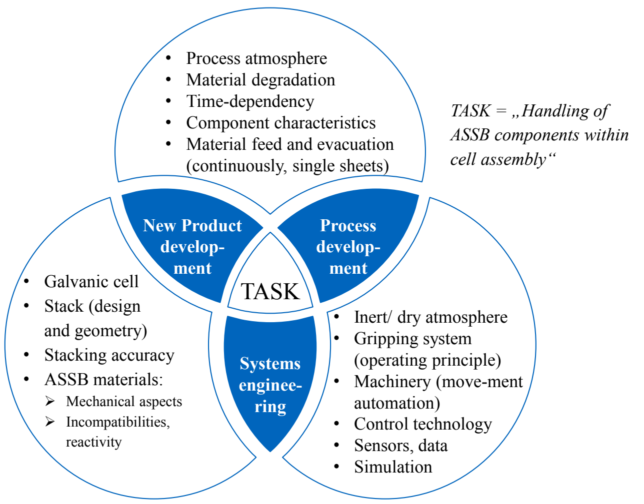

Fig. 2. Multi-disciplinary approach for handling ASSB cell components.

The definition of the material system of the ASSB galvanic cell represents a part of the product development. This allows to specifically define the respective components which will then be subject to handling operations during cell assembly. Another aspect of the product development is the definition of the cell and stack design.

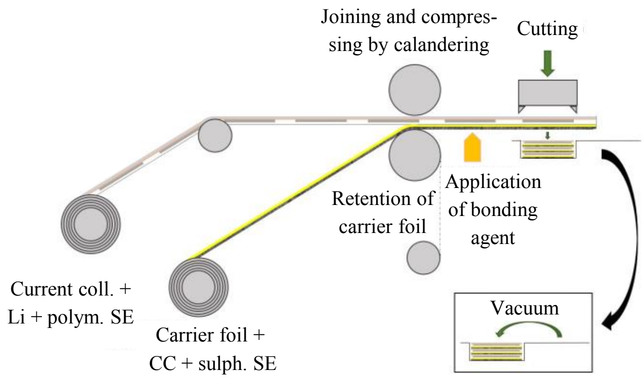

Fig. 3. Example for handling and processing components of a sulphidic ASSB during cell assembly.

This affects the systems engineering because it sets both the output parameters of the cell stacking process as well as its input parameters (components will possibly be fed in a different shape and/or arrangement for varying cell designs). Provided that a bipolar stacking concept is preferable for high energy density, the development of bipolar current collectors needs to be taken into account. The material transfer and feeding technology (roll-to-roll processes and subsequent cutting to size vs. handling and assembly of pre-cut single sheets and subsequent joining of layers) represents an intersection between product development and process engineering. Process development and systems engineering are innately closely interlinked.

The identification of handling processes is prerequisite for the conceptual design of the handling system for ASSB components. Consequently, the respective operating principles of grippers or conveyors that are required for handling the materials in ASSB cell assembly can be derived. This step, in turn, is strongly dependent on the characteristics of the components, e.g. feeding, handling and gripping systems will change depending on whether lithium is processed as a foil or on a substrate layer or whether individual components are fed.

The same applies to the processing of components as single layers in comparison to the processing of multilayer systems. As shown in Fig. 3, the feeding of different components from separate rolls, joining of multiple layers, and cutting of a multilayer system are directly linked to the cell stacking process.

5.2. Execution and evaluation of experiments

The design, construction and set up of a (prototype) handling system is prerequisite to carry out experiments and handling tests using a variety of samples. Thereupon, various experimental setups and test runs will be executed. A handling test rig (Fig. 4) that can be used as flexibly as possible for preliminary testing of handling the different components has been developed at the Institute for Machine Tools and Industrial Management. This handling test rig is used to investigate and compare different gripping principles for damage-free handling of single layers and multilayer systems of ASSB cell components.

The findings from trial runs using this handling test rig in different ambient conditions will prospectively allow to draw

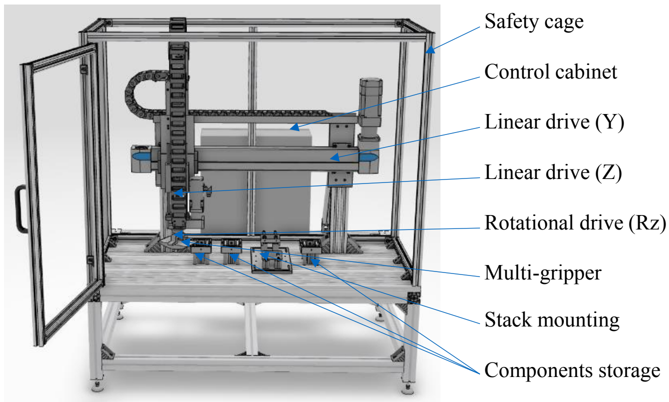

Fig. 4. Mechanical set-up of the handling test rig.

more profound conclusions and to define the specifications and requirements for automated handling and stacking of large format ASSB cell components. This will subsequently enable the development of an appropriate handling system for assembling ASSB cells on pilot scale.

5.3. Implementation of required environmental conditions

More profound knowledge regarding the required atmosphere for processing lithium, sulfidic SE and oxidic SE is required to ensure a sufficiently high product quality and safe handling and processing of the cell components. For this purpose, series of tests need to be carried out batch wise in different ambient conditions. If dry room conditions are not sufficient, the conception and design of a surrounding glove box is required, assuring an inert gas atmosphere (presumably argon), especially for handling and processing lithium metal.

Building on this the requirements for handling technology and machinery resulting from the special environmental conditions are derived. If possible, solutions from existing handling technology are adapted to the application. Otherwise a new handling solution (e.g. a specially developed gripping principle) must be developed for this specific application.

5.4. Fully automated feeding and stacking of components

As soon as the handling tests have shown positive results regarding functionality and the required environmental conditions have been realized, the automation of an integrated handling and stack assembly process will be the main focus of further investigations encompassing:

- integration of necessary sensor technology for control,
- optimization with regard to precision and repetitive accuracy to increase process yield,
- optimization of handling processes regarding speed,
- concept(s) for transport and feeding of components,
- prevention of cross-contamination,

- fixation, compression and evacuation of the cell stack,
- advanced system control (using a powerful PLC).

6. Conclusion and Outlook

Automating the cell assembly processes will be a prerequisite for mass commercialization of ASSB. A detailed mechanical characterization of lithium anodes, SE systems and compounds of SE and CC is necessary to specify the appropriate process conditions for handling the respective components during the assembly of ASSB cells. The integration of an appropriate manipulator into an all-enclosing glove box system will provide the technical basic framework that allows testing of the manufacturability of ASSB cell stacks under a controlled inert atmosphere. Hence, the results of this paper can assist in the transfer from laboratory to pilot and industrial scale for the production of All-Solid-State battery cells.

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