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All-solid-state lithium-ion and lithium metal batteries – paving the way to large-scale production

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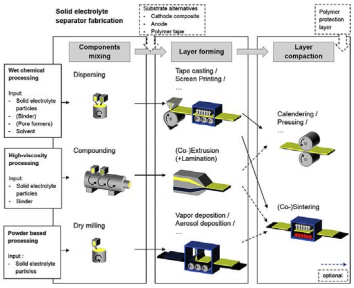
HIGHLIGHTS

- Requirements for industrial production of solid-state batteries are investigated.
- Process chains for electrode fabrication and cell assembly are presented.
- A detailed comparison with conventional lithium-ion cell production is given.
- Guidelines for stakeholders in the scale-up of fabrication are provided.

GRAPHICAL ABSTRACT



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ABSTRACT

Challenges and requirements for the large-scale production of all-solid-state lithium-ion and lithium metal batteries are herein evaluated via workshops with experts from renowned research institutes, material suppliers, and automotive manufacturers. Aiming to bridge the gap between materials research and industrial mass production, possible solutions for the production chains of sulfide and oxide based all-solid-state batteries from electrode fabrication to cell assembly and quality control are presented. Based on these findings, a detailed comparison of the production processes for a sulfide based all-solid-state battery with conventional lithium-ion cell production is given, showing that processes for composite electrode fabrication can be adapted with some effort, while the fabrication of the solid electrolyte separator layer and the integration of a lithium metal anode will require completely new processes. This work identifies the major steps towards mass production of all-solid-state batteries, giving insight into promising manufacturing technologies and helping stakeholders, such as machine engineering, cell producers, and original equipment manufacturers, to plan the next steps towards safer batteries with increased storage capacity.

1. Introduction

The need to store energy from renewable sources and the ongoing electrification of mobility increase the demand for safe batteries with high energy densities. Currently, lithium-ion batteries (LIB) are widespread and promising candidates for future application. Nonetheless, they suffer from raw materials availability, safety concerns, and limited energy storage capacity. State-of-the-art lithium-ion cells consist of two porous electrodes (anode and cathode) and a separator, as depicted in Fig. 1 (image a). The electrodes are coated onto a

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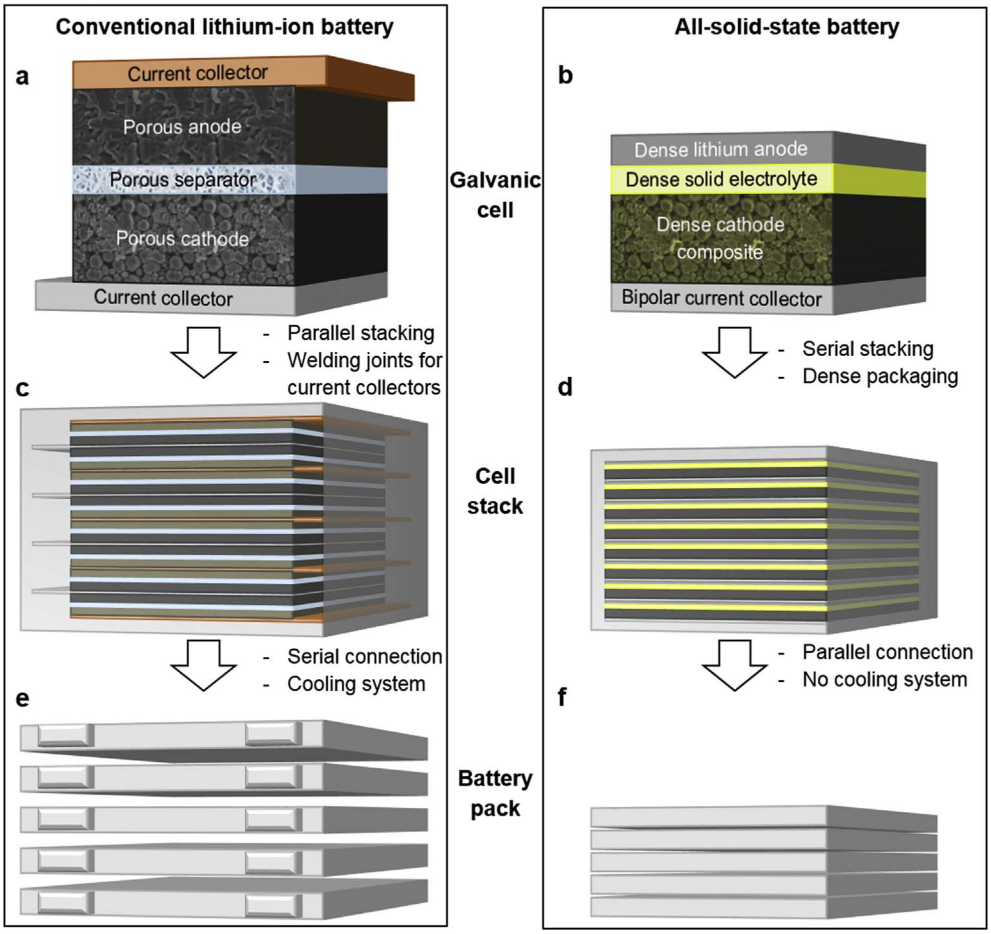

Fig. 1. Comparison of conventional lithium-ion battery and all-solid-state lithium battery at the cell, stack, and pack levels with potentials for increased energy density.

current collector, which consist of the active material, conductive agents, and binder [1]. The ion transfer requires a liquid electrolyte which is mainly composed of aprotic organic solvents and a conductive salt. Many of the issues that current LIBs are facing can be traced back to this liquid electrolyte. Safety concerns, in fact, arise from the flammability of the solvents [2]. Side reactions of the solvents and the conductive salt lead to capacity fading and aging [3]. During cell production, the cumbersome electrolyte filling and wetting process [4] as well as the extensive formation procedure contribute to high costs [5].

All solid-state batteries (ASSB), in contrast, are not only inherently safer due to the lack of flammable organic components, but also offer the potential for a dramatic improvement of energy density. Instead of a porous separator soaked with liquid electrolyte, ASSBs use a solid electrolyte, which acts as electrical insulator and ionic conductor at once (Fig. 1, image b). The use of a compact solid electrolyte acting as a physical barrier for lithium dendrites also enables the use of lithium metal as the anode material [6]. Therefore, an increase in volumetric energy density of up to 70% can be achieved compared to LIBs with conventional anode materials (e.g., graphite) [7]. Additionally, the electrochemical stability of some solid electrolytes facilitates the use of high-capacity (e.g., sulfur) or high-voltage (e.g., $LiNi_{0.5}Mn_{1.5}O_4$, LNMO) cathodes, which also leads to increased energy densities at the cell level [8–10]. In contrast to a liquid soaked LIB, a composite cathode is required for ASSBs, containing also solid electrolyte to create ionic pathways. In a conventional LIB, the liquid electrolyte interconnects all components of the battery cell. This leads to a parallel connection inside the cell stack (Fig. 1, image c). In contrast, in an ASSB the electrolyte is confined inside the galvanic cells. Thus, a bipolar stacking is facilitated, with the single cells connected in series by a lithium-ion isolating layer [11]. This can be used to increase the voltage of a battery cell and to reduce the amount of current collectors in the cell stack, as well as to optimize the packaging design (Fig. 1, image d). Finally, no cooling system is required for ASSBs due to the lack of flammable organic components, as depicted in Fig. 1 (image e) and Fig. 1 (image f). In fact, higher temperatures rather lead to an increased functionality due to increased conductivity [12].

The aforementioned advantages compared to conventional LIBs make ASSBs highly promising candidates for the application in electric vehicles and stationary applications. The implementation of ASSBs on the market will have to be accompanied by a scale up from laboratory research to industrial mass production. Currently, the powder pressing method is widely used to obtain dense solid electrolyte pellets. These, up to 1 mm thick, pellets are joined with cathode and anode layers and compressed to ensure mechanical contact [13]. However, a direct transfer of laboratory preparation methods to high volume fabrication processes on industrial scale is in many cases not possible. In contrast to polymer-based cells, where large-scale production has been successfully implemented in a similar fashion to conventional LIB production [14], only few publications actually describe attempts to fabricate large-format ASSBs with scalable production processes, such as wet coating [15,16], screen printing [17], and tape casting [18]. Theoretical considerations concerning large-scale manufacturing were presented by Hu [13] and Troy et al. [19]. An overview of current issues can be found in Kerman et al. [20], giving a detailed investigation on challenges from a product development perspective. The up-scaling of the materials, volume and mass reduction of inactive components for satisfactory energy densities, and implementation of scalable production processes will become the next big steps towards industrial fabrication of ASSBs [7,13]. Hence, this perspective manuscript contributes to paving the way to mass commercialization of ASSBs by investigating the multiple