

Application Fields of Extrusion Coating for Battery Manufacturing: A Mini Review

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ABSTRACT

For Lithium-Ion Battery (LIB) and future generation battery production, cost efficient and sustainable manufacturing routes need to be developed. A flexible design is a key aspect in order to be able to convert the production line for the fabrication of different material systems. This commentary is intended to show the ability of the extrusion coating to efficiently process solid polymer electrolyte separators as well as electrodes for LIBs.

Keywords: Extrusion coating; Flexible process design; Sustainable processing; Solid-state batteries; Lithium-Ion batteries

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INTRODUCTION

The extrusion based production route presented in Wiegmann and Helmers, et al. [1] enables further application fields which are discussed in the following. One of the fields of application is direct coating of the manufactured separator on the cathode for polymer electrolyte-based Solid-State Batteries (SSB). As stated in Wu, et al. [2] for SSB development next to enhancing the ionic conductivity a key challenge is the design of the solid interfaces. In contrast to the interfaces in conventional Lithium-Ion Batteries (LIBs) where the electrodes are wetted with a liquid electrolyte, for solid interfaces a sufficient contact needs to be generated during processing. If a free standing separator is manufactured and laminated onto the cathode, high interface resistances can occur limiting the electrochemical performance of the SSB.

In literature possibilities to enhance the contact in between the cathode and the Solid Electrolyte (SE) separator are currently explored. Ma, et al. [3] reduced the interfacial resistance in between the electrode and separator by *in-situ* polymerization of the separator film. Wang, et al. [4] used ultrasonic vibration to melt the solid polymer electrolyte and provide an enhanced contact at the interface.

Helmers, et al. [5] used a lamination step at 0.976 MPa and 90°C to provide a sufficient contact in between the separator and the cathode. These methods result in an additional process step with related investment and operational costs. The process presented in Wiegmann and Helmers, et al. [1] offers the opportunity of direct casting of the solid electrolyte separator on the cathode. Thereby, the molten state of the SE is used to achieve an improved wetting of the cathode. Hence, with the developed process route currently heavily investigated interface challenges can be approached without adding further process steps.

LITERATURE REVIEW

Next to that, a current research topic is the flexible design of production lines and providing the possibility of converting the process route for the application on different material systems. Therefore, a possible transfer of the developed process route to the manufacturing of conventional LIBs was evaluated. Lithium-Ion Battery manufacturing aims to reduce CO₂ emissions and process costs by developing innovative and scalable electrode production methods. Direct coating of electrode layers *via* an extruder enables the processing of increased solid fractions, thus saving drying costs and time.

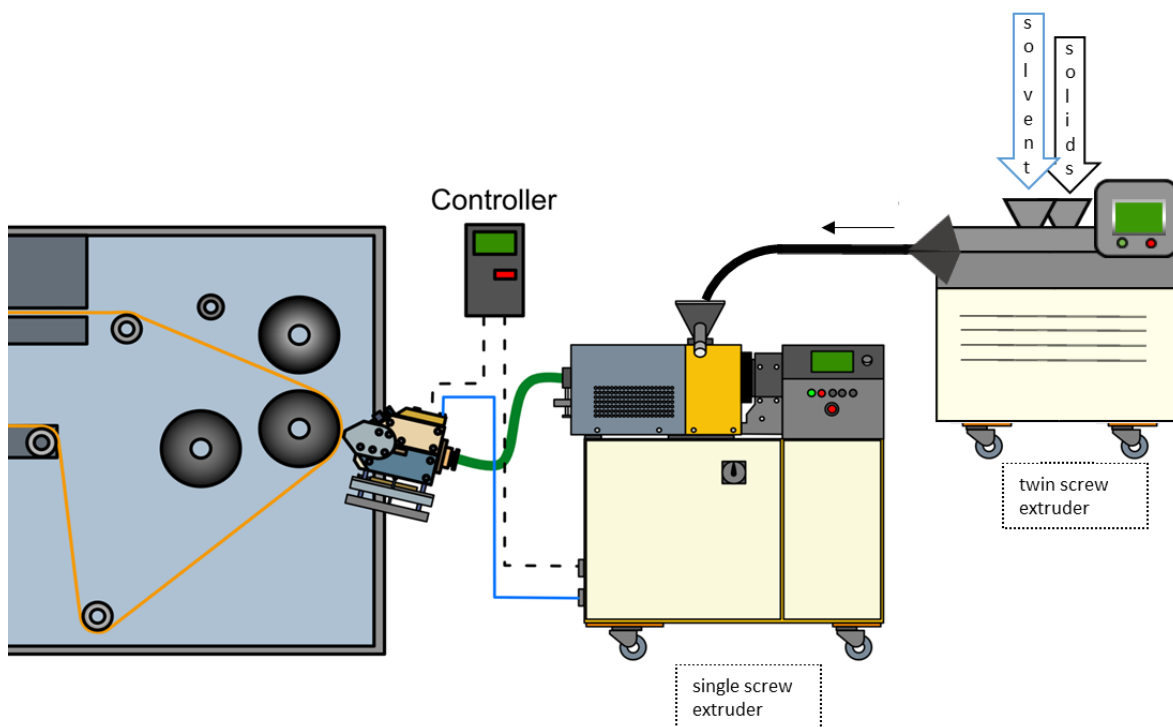
Conventional production of electrodes, on the other hand, is mainly based on low-viscosity wet processing. Typically, relatively low viscosity anode pastes (about 50% solids content) and cathode pastes (about 70% solids content, mostly NMP/NEP-based) are produced [6]. The pastes are applied to the current-conducting films using a comma-bar or slot die coating process. Using convective drying, the electrode layer is solidified by evaporation of the solvent. This process correlates with long drying times and associated high energy and investment costs [7].

In addition, the segregation of inactive electrode components of lower specific density can be observed in this form of electrode processing due to the high solvent content, which affects their mechanical properties and charge transfer capability [8,9]. The established batch mixing methods also bring various disadvantages besides mixing time, such as quality variation between different batches, many manual steps, and cleaning efforts.

To address these challenges and drawbacks, the process developed in "Highly scalable and solvent-free fabrication of a solid polymer electrolyte separator *via* film casting technology" [4] was adapted to lithium-ion electrode processing. This adapted process concept is shown schematically in the following Figure 1.

As visualized in Figure 1, the developed process operates continuously using extrusion technologies. Extrusion technologies can handle higher solids content without allowing dead zones to remain during mixing. The dispersing twin screw extrusion technology is combined with a single screw extruder, used for direct lamination respectively coating of electrode slurries. The pastes within the single screw extruder, characterized by higher solids contents, can be conveyed and pressed through a slot die in order to be directly laminated onto a substrate foil (copper or aluminium).

Figure 1. Process concept of extrusion coating for manufacturing of Lithium-Ion-Batteries.



Increasing solids content (approximately relative increase of 20%) within electrode fabrication significantly reduces drying energy. As the cumulative energy input is dominated by the drying process, modifications in the solvent content of the slurry will help to minimize it.

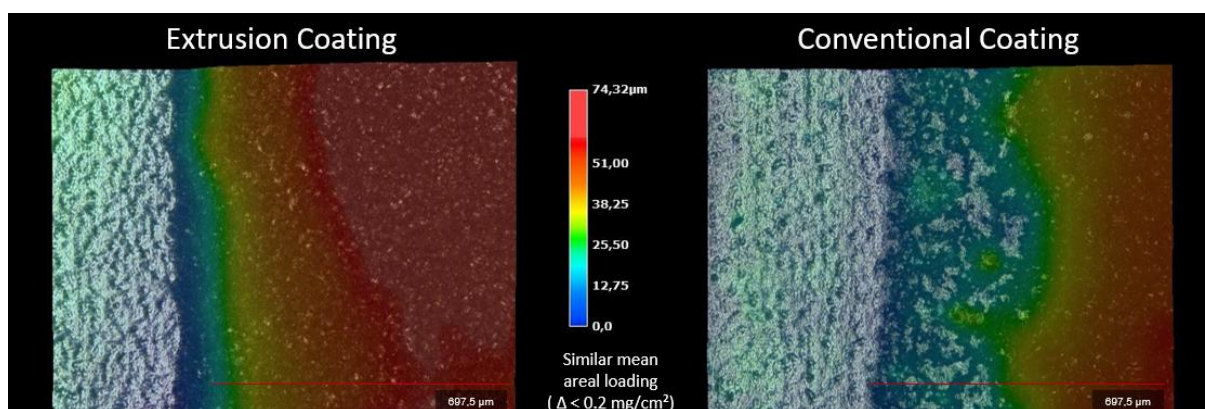
By reducing the electrode's solvent share by about 20%, drying time could almost be halved. This allows for faster processing speeds and higher throughput during the coating and drying step, while reducing the risk of binder and conductive additive migration to the electrode surface. Adapted drying profiles could represent a further option in order to make battery production more energy-efficient.

DISCUSSION

At the same time, the continuous mixing system based on the twin-screw extruder can save 60% of the process-specific mixing energy and a significant increase in throughput per machine unit compared with batch mixing. Further, the high solids content (high viscosity pastes) can increase throughput by maximizing the plant-specific mixing/coating throughput. In addition, increased solids content enables the formation of dimensionally stable edges.

In contrast, edges of low viscosity manufactured electrodes tend to flow (swell) towards the substrate edges (heavy edges) after coating application [10]. In Figure 2, 3D-microscopic pictures for different coating processes are compared at 50% solids content.

Figure 2. Edge formation during extrusion coating and conventional coating.

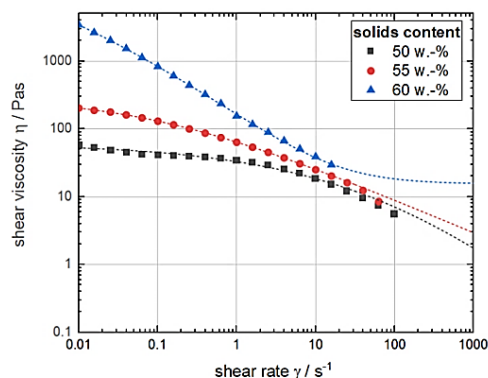


For quasi same areal loading and same pre-processing of the electrode slurry, well-defined edges are recognizable during extrusion coating while for conventional coating, coating islands can be observed before a continuous film appears. It can be expected that, by increasing the solids content, sharper edges might be obtained since the slurry would spread even less at higher viscosities. According to Spiegel, et al. [10] by selecting the appropriate shim geometry, edge elevations can be further improved.

Increasing the solids content of an electrode slurry, for example by increasing the active material or the conductive additive shares, will significantly change its structural properties which can be challenging to process but will lead to a strongly reduced energy demand. This depends on several factors such as particle size, solvent, binder and their interactions within the slurry. The conductive agent should be well distributed and dispersed within the mixture to effectively build conductive paths or networks within the electrode. The conductive additive particles are very small, likely to agglomerate and therefore, tend to immobilize solvent which can lead to higher viscosities and, furthermore, inhomogeneous distribution. Accordingly, when less solvent is used, it is even more important to reach a good dispersion, mainly to overcome viscosity and, therefore, flow issues. Consequently, to enable for higher solvent mobility, agglomerates have to be broken up into aggregates. This can be achieved by mechanical stressing of the slurry which is implemented *via* kneading zones within the twin-screw extruder.

Exemplary viscosities of the slurries processed *via* twin-screw extrusion at different solids contents are shown in Figure 3.

Figure 3. Viscosities of coated and adapted slurries at different solids contents. Note: ■ 50 w%, ● 55 w%, ▲ 60 w%.



In Figure 3, the viscosities of slurries with different solids contents are depicted at increasing shear rates. Hereby, the shear-thinning behaviour of the electrode slurry becomes apparent and for the highest solids content, a higher inclination can be observed when turning to higher shear rates. Viscosities of all depicted slurries converge at high shear rates, leading to the conclusion that there might be a “sweet spot” where they can still be coated in very similar manner. Shear rates that typically occur for slot die coating are shown [11]. By extrusion coating, these high shear rates can be reached at almost no pulsation which represents a big benefit of the presented process. A direct connection of the twin-screw extruder with the slot die would lead to strong pulsation effects. Hence, the possibility of low-pulsation conveying at high viscosities for the extrusion coating will be further investigated in upcoming studies.

To further tweak viscosity after slurry production, both slot die and single-screw extruder can also be heated by the same heating unit. By raising the temperature, the kinetic energy of the material molecules within the suspension can be increased, consequently it will flow more easily and in this manner, viscosity may be adjusted within the operating range of the slot-die as observed from Hawley, et al. [12]. The results presented emphasize the extended application possibilities of the developed process route. Hence, deeper investigations regarding the extrusion coating and direct coating should be conducted.

CONCLUSION

Summarizing, a process concept was introduced that offers cost efficient and sustainable processing not only for future generation battery systems as presented, but also for conventional LIB manufacturing. For SSB production the direct coating of the separator on the cathode can reduce interfacial resistances, which is currently an area of intense research. Besides, the fabrication route can provide processing of low-solvent content and accordingly, highly viscous slurries which enables for significant reduction of the cumulative energy demand during electrode and battery cell production for conventional LIB's. This shows the capability of the process to dramatically decrease CO₂ emissions.

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