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Benefits of solvent-reduced twin-screw compounding for cost-efficient, eco-friendly, and high-performing lithium-ion batteries

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Challenges for efficient battery production

The global production of lithium-ion batteries will increase enormously with the increasing demand for electromobility and energy storage. In this respect, ecologically and economically efficient production of electrodes is crucial. 10 % of the greenhouse gas emissions in battery production yields from electrode coating and drying.[1] Conventional coating of the electrode collector foil requires low-viscous slurries with a solvent content of 45 %.[2] Currently, the reproductive toxin NMP (N-methyl-2-pyrrolidon) is used as a solvent in cathode production and needs to be recovered and recycled. The drying and solvent recovery of conventionally wet electrode slurries after coating onto current collector foil accounts for the major amount of energy consumption (approximately 45 %)[3] in lithium-ion battery manufacturing. The drying ovens, which can be up to 80 meters long, occupy a substantial amount of plant space.

A promising approach to achieve more cost and energy-efficient battery production is to develop innovative processes for dry or low-solvent electrode manufacturing utilizing continuous twin-screw compounding. It bears the potential for significant solvent reduction while maintaining a high dispersion quality of the electrode pastes.[4] Furthermore, the adaptable screw design can help to optimize electrode structure leading to performance enhancement.

Low solvent battery paste compounding

Twin-screw extruders achieve fine dispersion in high-viscous pastes through strong shear forces acting on the material. The small distance between the extruder screws and the barrel yields higher mass-specific-power input than planetary mixers allowing the reduction of solvent content by 50 % in cathode pastes.[4] The segmented screw design enables the optimization of the mixing process to achieve optimal morphology and surface area, which correlates with enhanced rate capability during cycling.

Wiegmann et al. 2023[5,6] investigated solvent-reduced graphite anodes and lithium ferro-phosphate (LFP) cathodes with polytetrafluoroethylene (PTFE) mixed in a twin-screw extruder with different screw configurations (i.e., **configuration 1**: five kneading blocks, **configuration 2**: three kneading blocks, and **configuration 3**: no kneading blocks (Figure 1)) and compared those to wet-processed electrodes mixed in a conventional planetary mixer as a reference.

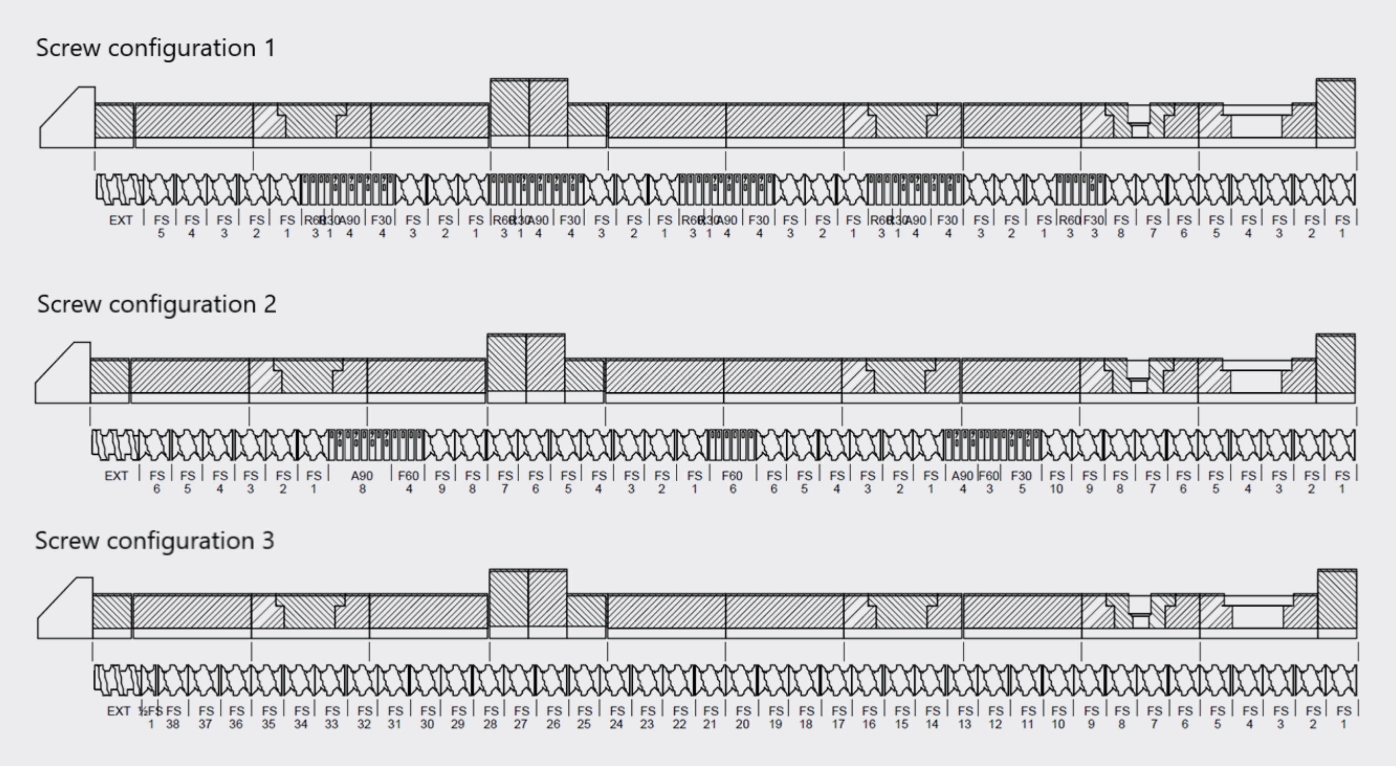

Figure 1: Parallel twin-screw extruder screw configuration 1 with five blocks of kneading elements, screw configuration 2 with three blocks of kneading elements, and screw configuration 3 with no kneading elements. (Adapted from Wiegmann et al. 2023).[5]

Figure 2 shows the specific power input of the extruder equipped with the screw configurations in Figure 1 and the planetary mixer as the reference. The specific power input correlates linearly with the torque applied onto the screws in the extruder or the rotor in the planetary mixer. In principle the torque increases with the number of kneading blocks. However, a higher torque and therefore a higher specific power input was measured in the extruder with the screw configuration without kneading blocks. This is presumably due to insufficient break-up of particle agglomerates that increase the anode paste viscosity.[5] This emphasizes that the extruder screw configuration plays an important role in the dispersion quality and needs to be designed with great care.

Using twin-screw extruders for electrode processing

Thermo Fisher Scientific offers a range of twin-screw extruders that are optimized for battery electrode mixing processes with 11 mm, 16 mm, or 24 mm screw diameters. All instrument sizes share the split-barrel design and the segmented screws that allow for fast cleaning and process customization (Figure 2). This renders them ideal for development of novel formulations and evaluation of extrusion in lab and pilot scale as a compounding solution for production.

The Thermo Scientific™ Energy 11 Twin-Screw Extruder is a bench-top model with 11 mm screw diameter and can handle throughputs of dry battery pastes starting from 200 g/h (Figure 3). This instrument size is designed for early trials when expensive materials need to be used in lower amounts or if numerous different formulations need to be processed in less time.

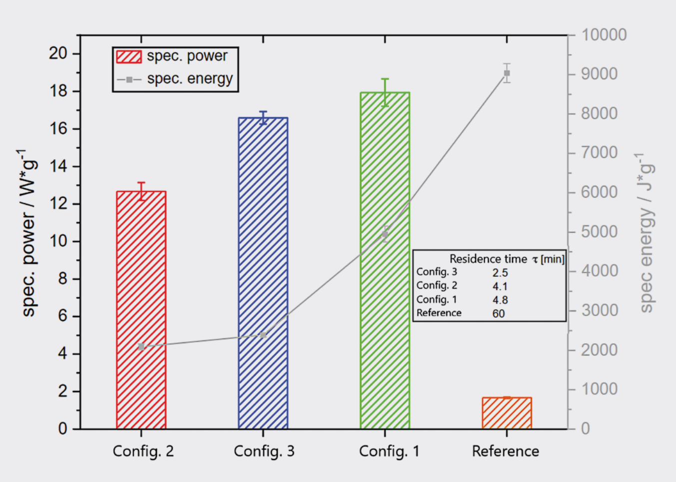

Figure 2: Specific power input and specific energy input (i.e., product of specific power and residence time) during mixing of semi-dry graphite anode pastes in an extruder with two different screw setups and a wet graphite anode mixed in a planetary mixer as reference. (Adapted from Wiegmann et al. 2023).[5]

Figure 3: Split barrel design of the Energy 11 Twin-Screw Extruder.

Increasing the screw diameter of an extruder also increases the output of electrode material per unit of time. Depending on the availability of material, electrode pastes can be compounded with throughputs up to 30 kg/h on Thermo Scientific Twin-Screw Extruders with 24 mm screw diameter.

All extruders are available in stainless steel grades that are chemically resistant against corrosion, in CPM hardened steel withstanding abrasion, or in nitriding steel 1.7361 (EN40B) exhibiting a well-balanced mix of both qualities.

The segmented screw design and the individual screw elements that determine the screw configuration are key factors in the individual process design for optimizing mixing properties. The screws are assembled using individual screw elements (see Figure 4) which are put onto hexagonal shafts. This design allows for alternating sections of material transportation, utilizing conveying elements, and material mixing, employing kneading elements.

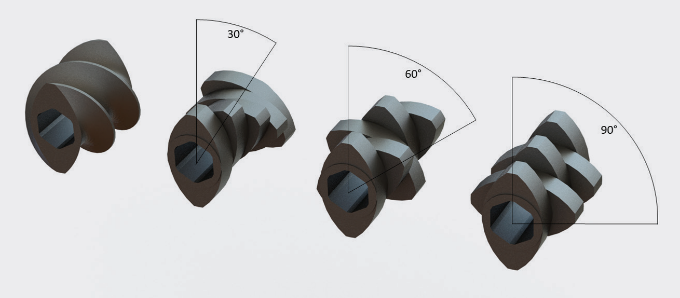

Figure 4: From left to right: conveying element, 30° kneading block, 60° kneading block, 90° kneading block.

Additionally, conveying elements introduce a certain shear energy, as their tips have a very small clearance to the barrel walls. To introduce higher shear energy, a mixing block, consisting of individual kneading elements, is necessary. The kneading elements can be arranged with different offset angles between the individual elements such as 30°, 60°, and 90°. The larger the offset, the greater the introduced shear energy, which enables high dispersive mixing.

The ability to optimize these mixing blocks in terms of their length, number, and shear energy input (through the offset angle) allows for finding the optimal distributive and dispersive mixing for a given electrode formula.

Optimizing electrode processing

The HAAKE™ Rheomex PTW 16 OS (16 mm screw diameter) displayed in Figure 5, is successfully used in research projects for innovative electrode manufacturing.[5,6]

The twin-screw extruder compounds anode or cathode material with minimal solvent addition. The highly viscous pastes are processed into pellets with the FaceCut 16 Pelletizer (Figure 6). In this shape they are easily transported and stored without risk of sedimentation. To form electrodes, the pellets can later be coated onto collector foil and calendared in one step.

Figure 5: HAAKE Rheomex PTW 16 OS Twin-Screw Extruder with 16 mm screw diameter.

Figure 6: FaceCut 16 Pelletizer (left) cuts extruded paste into pellets (right) with rotating blades directly at the die exit.

This electrode manufacturing route is scalable to mass production and prospectively demands 60 % less energy than conventional manufacturing.[5] Scientifically based strategies for scale-up of extrusion processes have been successfully applied, also in other industries.[7] The same strategies can be used when analyzing the critical quality attributes of the electrode structure and correlating them to extrusion process parameters.

Within this quoted work, extruders with identical geometries have been used. This makes scalability of the compounding process between different extruder sizes easier, e.g., the free volume stays constant (same ratio between inner to outer screw diameter).

Effects of extrusion screw configuration on the electrode structure and performance

The mixing technology, screw configuration, and specific power input applied during electrode mixing affect the electrode structure, as demonstrated by the decrease in pore diameter observed in coated anodes produced using a planetary mixer and a twin-screw extruder (refer to Figure 7). With an increase in shear and the corresponding power input applied by the extruder, the pores in the coated anodes become smaller, resulting in an increase in specific surface area.[5] The shear that the material experiences is determined by the number and configuration of the kneading blocks on the extruder screws, as well as processing parameters such as screw speed and material throughput. The resulting electrode structure, characterized by such factors as the specific surface area (i.e., the surface area per unit mass), plays an important role in determining its ionic resistance, charge-discharge performance, and cycling stability. The specific surface area of the anodes produced with the extruder is orders of magnitude higher than that of anodes mixed with the planetary mixer.

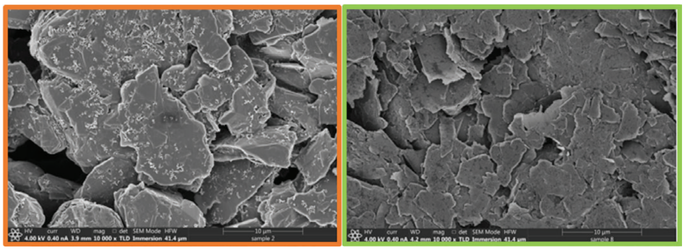

Figure 7: Scanning electron microscope images of cross-sections of the anodes mixed with the planetary mixer (left) and the extruder with screw configuration 1 (right) (Adapted from Wiegmann et al. 2023).[5]

The discharge capacity of full cells with the anodes produced using the planetary mixer as a reference and the extruder with different screw configurations is shown in Figure 8. The ionic resistance of an electrode decreases with the increase of the specific surface area to some extent,[5] and this enhances the rate capability during cycling as visible in Figure 8. Full cells with anodes produced using a twin-screw extruder, with screw configuration 2 and with moderate shear exhibits significantly higher discharge capacity than those with anodes mixed in a planetary mixer up to a C-rate of 2 C. However, the beneficial effect inverts above a certain specific surface area of the anode. Beyond that, excessively high surface area in contact with electrolyte results in an undesirable large solid-electrolyte interface (SEI).[5] The excessive SEI presents a barrier for ionic transport increasing the ionic resistance and lowering the discharge capacity as shown in Figure 8. Hence, mixing the anode in the extruder with a screw configuration applying moderate shear and increases the specific surface area to an optimum, improves the cell capacity.

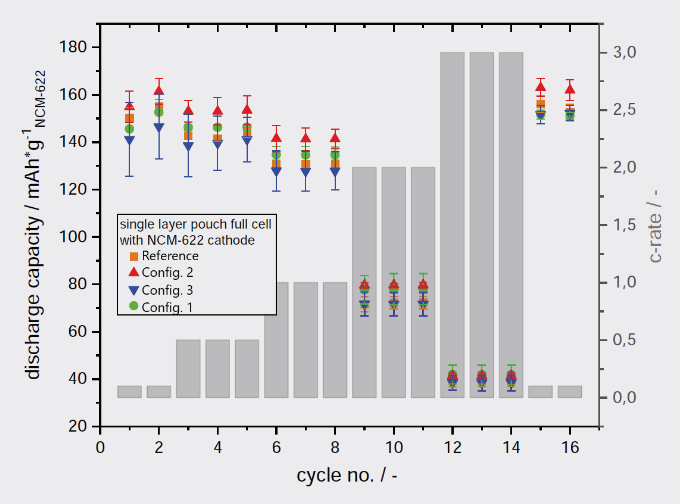

Figure 8: Discharge capacity of full cells in dependency of different C-rates for the different anode processing routes. (Adapted from Wiegmann et al. 2023).[5]

For solvent-reduced and dry cathode processing, PTFE was proven as a suitable binder when mixed in small amounts of 1–2 % with the cathode active material under high shear.[3] It forms fibrils that bind the particles together and gives the cathode mixture elasticity and mechanical strength so that it can be calendered into crack-free cathode coatings. The extent of fibrillation depends not only on the PTFE grade but also on the power input during the mixing process.

Figure 9 shows scanning electron microscope images of LFP cathode mixes with PTFE fibrils produced in the extruder with screw configurations 1 and 3, with zero and 5 mixing blocks, respectively. The higher power input from the mixing blocks results in a higher quantity of fibrils and the absence of fibrils thicker than 1 µm. The latter are visible in the cathode mix produced with the screw configuration without mixing blocks. A more homogenous distribution of smaller fibrils favors the electrode elasticity and adhesion to the current collector. However, PTFE is nonconductive, and its addition reduces the cathode electric conductivity by orders of magnitude. This effect is more pronounced for cathodes mixed with the screw configuration with five mixing blocks, presumably because the higher number of smaller fibrils isolates more electric conductive pathways.[6]

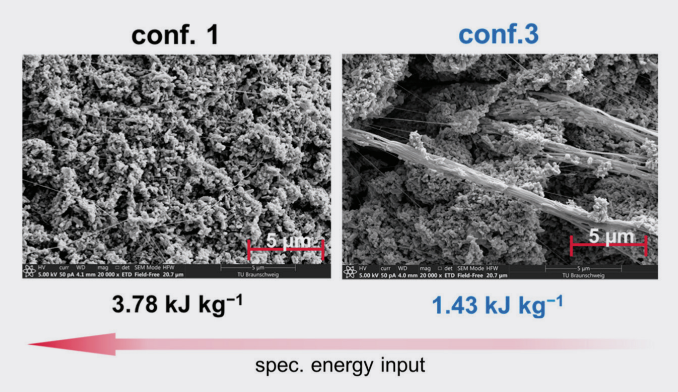

Figure 9: Scanning electron microscopy images of granules in LFP cathode pastes with PTFE fibrils mixed in the extruder with screw configuration 1: five kneading blocks (left) and screw configuration 3: no kneading blocks (right). The specific energy input of the mixing process is stated below, respectively. (Adapted from Wiegmann et al. 2023).[6]

The differences in PTFE fibril number, size, and distribution also affect the cathode performance as can be seen from the specific discharge capacities of half cells in Figure 10. The discharge of the reference cathode and the cathode mixed with screw configuration 3 is close to the theoretically calculated capacity value of LFP (155 mAh g^{-1}) at low cycling rate of 0.1. At cycling rates larger than 0.5, the capacity of the cathode mixed with screw configuration 3 exceeds the reference. Wiegman et al.[6] blame the ionic conductivity decreased by the binder within the reference for the drop in capacity at larger cycling rates. The cathode mixed with screw configuration 1 with five kneading blocks shows the lowest capacity regardless of the cycling rate. The latter can be rationalized by the lower electrical conductivity of the cathodes with the homogeneous distribution of fine PTFE fibrils. The highest cycling stability is also exhibited by the cathode mixed with the extruder with screw configuration 3, as can be seen in Figure 10. It shows the lowest capacity loss after 40 charge/discharge cycles. To achieve the desired mechanical properties and prevent an excess of fine fibrils that could reduce electrode conductivity, it is necessary to carefully tune the PTFE fibrillation during the mixing of dry and solvent-reduced cathodes, ensuring sufficient distribution without exceeding the required amount. The optimization of the specific power input required for fibrillation can be done precisely via the extruder screw design.

Conclusion and outlook

These results emphasize that twin-screw extrusion can present a beneficial electrode mixing technology that helps to overcome challenges present in today's wet processing manufacturing routes.

- Reduced overall processing time
- Consumption of harmful solvents can be dramatically reduced
- Energy for the drying and recycling process is significantly reduced
- Segmented screw design enables tailored mixing processes
- A scientific scale-up approach is available
- Dry electrode pellets can be stored without phase separation

These results also demonstrate the importance of a well-balanced specific power input to enhance battery performance. Therefore, tailoring the extrusion process involves identifying the optimal screw configuration and processing parameters. A small lab-scale twin-screw extruder serves as a versatile tool for accomplishing this objective.

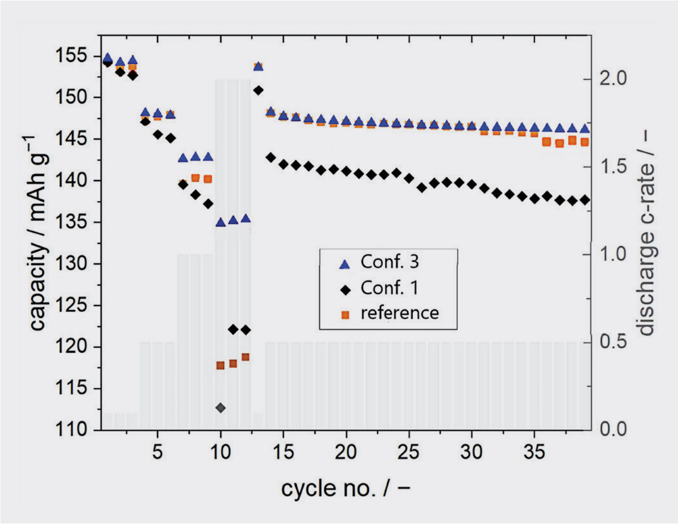

Figure 10: Specific discharge capacities of half cells in dependency of different C-rates for the different cathodes mixed in the extruder with screw configuration 1: five kneading blocks and screw configuration 3: no kneading blocks, and for the wet processed cathode mixed with planetary mixer. (Adapted from Wiegmann et al. 2023).[6]

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