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A Review of Process Innovations in the Cell Finishing of Lithium-Ion Batteries in Large-Scale Production

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Abstract

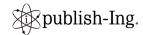
The European Union's ambitious climate targets will make climate-friendly storage technologies essential. More than any other, this decade could be marked by battery technology, especially the lithium-ion battery (LIB). In addition, various trends in mobility and consumer electronics are spurring the cross-industry use of this secondary storage device. As a result, the need for additional production capacities is rising, and the need for vertical integration of the value chain of LIB in Europe. In current forecasts, Europe has a considerable deficit between battery cell demand and production capacities. The deficit highlights the need for additional capacities and effort to develop new production systems. Furthermore, production technologies remain challenging, as high reject rates are expected initially, and a reduction of costs at the battery cell level is mandatory. Formation and aging as part of the cell finishing are the production steps with the highest processing time and space requirements. The formation can take up to 24 hours, and the subsequent aging between 8 to 36 days. It thus represents the biggest bottleneck. In large-scale production, various process innovations are being worked on, depending on the degree of automation. However, a systematic study of the impact of these process innovations is hardly ever carried out. Various approaches are conceivable here: Innovative formation protocols, optimized plant technology, flexible goods carrier systems and other process-related innovations. This paper provides researchers and industry experts with meaningful insights into the status quo and future developments in the cell finishing of battery cells through a comprehensive research approach. These trends will be presented and systematically evaluated to identify the most significant levers to reduce costs and time. It reviews process innovations in cell finishing to approach this research gap and aims to answer how these innovations will benefit and shape the large-scale production of lithium-ion battery cells.

Keywords

Lithium-Ion Battery Cell Production, Cell Finishing, Cell Conditioning, Formation, Soaking, 2nd Filling, Aging, Process Innovations, Large Scale Production

1. Introduction

Lithium-ion batteries are one of the key technologies for the transition from fossil fuels to renewable energies and thus have an outstanding significance on the way to a world free of pollutant emissions and for the mitigation of climate change. [1] Their ability to store electrical energy almost without a loss will make it possible to switch electrical energy production to renewable sources in the long term. Due to their technical properties as electrical energy storage devices, lithium-ion batteries have a wide range of applications. They are, for example, the most frequently used energy storage device in mobile electronic devices. [2]



Technological progress and announcements by Original Equipment Manufacturers (OEMs) about a fully electrified portfolio, among other things, are leading to a rapid increase in demand for lithium-ion batteries and massive investments in battery development and production [3,4]. The production of lithium-ion batteries is very time-consuming and cost-intensive and can still be considered challenging regarding energy consumption. Studies [5,6,7,8] show that especially the formation accounts for a great share of the operational cost and total energy consumption per year within battery cell production. The formation is one of the central stages in the production of lithium-ion battery cells. It involves the initial charging and discharging of the manufactured cells and influences their electrochemical performance throughout their lifetime by ensuring the formation of a protective layer that prevents cell decomposition reactions between anode and electrolyte. [9]

2. Overview of the Battery Production Processes

The materials and cell chemistry (for the anode and cathode) of a battery cell must be defined as well as the cell format. As a first step, these parameters must be determined. Product specifications have far-reaching implications, especially for battery cells for production processes. After that, the resulting technology chains can be examined in more detail. This review will focus on the prismatic cell with conventional cell chemistry and dimensions to narrow down the innovation scope (Figure 1).

The overall process of battery cell production can be divided into three stages: Electrode production, cell assembly and cell finishing (Figure 1). The individual process steps may vary depending on the cell format and the materials selected, but the basic sequence is essentially identical for all common lithium-ion batteries. Starting with electrode production, the raw materials are first processed into anode and cathode. These are then assembled into battery cells together with a separator, electrolyte and housing in cell assembly. In the last production step of cell finishing, the manufactured cells are formed and aged, followed by quality control (or characterization), which is also performed during aging. Afterwards, the battery cells are ready to be graded, packed, and shipped.

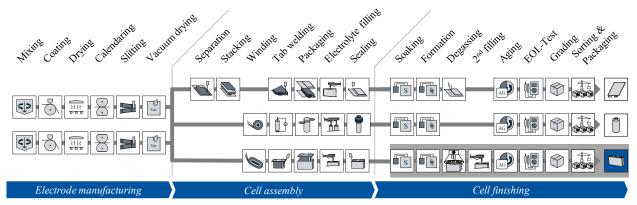


Figure 1: Overview of the process steps in the production of lithium-ion battery cells with different formats [10,11]

Figure 1 provides an overview of the battery cell production process chain. In electrode production, there are two lines of the same configuration each for anode and cathode. In cell assembly, the process steps depend on the cell format and design. The focus of this paper relates to the area of cell finishing. Due to long process times of up to 7-28 days in total, electrolyte wetting and the subsequent process steps of formation and aging in particular are major bottlenecks in battery production. The process sequence is variable and product dependent. [12,13,14]

This paper explores process innovations in cell finishing to classify different approaches and opportunities for optimization to address this bottleneck. The objective is to provide insights into how different process innovations in cell finishing will promote and shape the large-scale production of lithium-ion battery cells.

3. Process Chain in the Cell Finishing

Cell finishing is the last stage in the production of lithium-ion batteries. The battery cells are fully assembled. The cells are now electrochemically activated. Depending on the finishing protocol, the battery cells pass through the process steps and measurements in a different order. [10,11]

3.1 Soaking

During the soaking process, the cells are stored in a high-temperature environment to reduce the viscosity of their electrolyte and thereby ensure its homogeneous distribution in the cell. The required duration of the process at a given temperature is highly dependent on the size of the cell, its format, the cell chemistry, and the electrolyte-filling process [15]. It is common to consider the process as finished without validating its successful completion since the direct determination of the homogeneous electrolyte distribution requires complex measurement procedures [12,16].

Innovations

One option to indirectly determine the wetting degree of the cells is electrical internal resistance measurements [17,18]. Such measurements have the advantage of being very quick and they do not require the cells to be removed from their trays. Furthermore, no additional measuring equipment is required since both direct current (DC) and alternating current (AC) internal resistance are part of the regular quality control measurements. However, since both measurements do not take place within the soaking units, additional transport routes and logistical challenges arise. In addition, the validity of these indirect measurements is significantly lower compared to a direct analysis of the wetting degree, as several other factors influence the results. Therefore, resistance measurements are primarily advantageous if they are performed several times so that the interfering factors can be subtracted, and the actual course of the electrolyte distribution can be approximated. [19]

Evaluation

The implementation of electrical resistance measurements during the soaking process is a simple method for an indirect evaluation of the process progress. However, due to the additional transport distances and measurement procedures, it is only economical if the processing time saved exceeds the costs for additional measuring equipment or additional transport routes. Therefore, it is advantageous for large cells that would require an exceptionally long soaking time. The applicability for large-scale production with fully occupied transport systems is therefore limited. But the future potential can be significant if methods are found by which a single measurement during pre-forming is sufficient to analyze cell-specific the required soaking time. The foundation can be artificial intelligence (AI) methods or extensive knowledge about analyzed cells. In comparison, non-electrical methods for determining process progress typically require the removal of cells from their trays or time-consuming measurements. The implementation of such methods is therefore even more costly but offers a comparable acceleration potential. [17,20]

3.2 Formation

Formation represents the processes of cell finishing in which the manufactured and soaked battery cells are charged and discharged for the first time and thereby activated. However, there are several key differences in comparison to the regular charging process, which must be considered when designing a formation protocol. The primary objective of the formation is to cause the controlled development of passivating boundary layers on the surfaces of the electrodes. [12,21,22,23]

The traditional method to avoid damaging reactions within the cell during formation is to charge and discharge at low C rates between their upper and lower voltage thresholds for at least two full cycles [21,22]. However, this results in the process taking up to 48 hours. Through a detailed analysis of the reactions leading to a sufficient solid electrolyte interphase (SEI) and the deleterious effects that may occur, accelerated formation procedures can be applied.

Innovations

The easiest way to implement accelerated formation protocols is to use adjusted current and voltage thresholds. This method has the advantage that no increased requirements are imposed on the equipment. A typical way to speed up the process is to increase the current during the discharge phase [23]. This can

prevent the damaging reactions at the anode that occur primarily during charging, while at the same time accelerating the process. Further acceleration can be achieved by not fully discharging the cells. This takes advantage of the fact that the functionally more important inorganic compounds in the SEI are formed primarily in the high cell voltage range [22,23].

In the so-called dual-current formation procedure, the charging power is adjusted in two stages depending on the cell voltage. As in a traditional formation procedure, the charging process starts with a low C rate in the range of 0.1 C. Once the cell voltage exceeds a predetermined value, the charging rate is increased abruptly to a value that is typically around 70 % of the C-rate capability of the cell. Thereby, there is sufficient time for the formation reactions of the organic SEI compounds to occur. Another positive effect is that the increased charge rates accelerate the formation of inorganic SEI compounds in the upper cell voltage range. However, many damaging reactions are also driven by the combination of high charge rates and high cell voltages. Therefore, a decision on the timing of the change and the increased charge rate must be balanced between the demand for acceleration and the risk of damage. [23,24,25]

It can be useful to perform part of the formation before soaking or between two soaking sub-processes. This additional formation step, which aims to set the cell voltage before the soaking, is usually called pre-charge. By charging to about 2.5 V, decomposition effects that occur after a certain time between the uncharged anode and the copper foil can be avoided. This allows the soaking process to be carried out for a longer time, which is especially important for large format cells. During the pre-formation, charging must be conducted at particularly low C rates of about 0.05 C due to the uncompleted electrolyte distribution. By increasing the targeted voltage of the pre-formation to around 3.3 V, the increased cell temperature during soaking can be utilized to initiate and accelerate initial SEI-forming reactions. After storage above 40 °C and at a cell voltage of at least 3.3 V for 24 hours, a sizable portion of the organic compounds has already formed in the SEI. Thus, the subsequent main formation can start with higher C-rates without negative influences on the cell. [23,25]

Evaluation

The main advantage of any alternative formation procedure based solely on adjusted thresholds for current and voltage is that it is quite simple to implement in any existing cell finishing line. The only condition that such methods impose on the equipment is that it must be capable of delivering the desired current. At the same time, such methods provide a time-saving potential since typically a large part of the formation time takes place in unnecessary potential ranges. Therefore, it can be concluded that these methods offer a good mix of applicability for large-scale production and acceleration potential. [22,23]

Besides simply adjusting the threshold values, several other electrical methods have the potential to accelerate the formation process. However, these often place special demands on measurement electronics and power electronics. The possibility of subsequently integrating them into an existing series production can therefore be limited. An even greater acceleration potential can be achieved by using a dual-current procedure. But the applicability of this process depends very much on the cell chemistry and requires power electronics that can provide the high currents needed during the second charging phase. [23,24,25]

A major disadvantage of pre-charging into the SEI forming potential range is that this acceleration must be substituted with an extended pre-charge phase. In addition, a high degree of wetting of the electrodes with electrolyte should already be present before the pre-charge to prevent inhomogeneous SEI formation. Therefore, the overall acceleration potential of this technology is limited, but a positive influence on cell performance can be observed while at the same time being easily applicable in large-scale production. [23,25]

3.3 Degassing

During formation, some components of the electrolyte are reduced [26,27]. The resulting gases accumulate inside the battery cell, as these are already sealed. In hardcase cells, the pressure rises due to gas buildup inside the cell. For safety and quality reasons, the gas is therefore vented from the cell in the degassing process step, which is usually integrated as a separate station in fully automatic finishing lines [28]. In prismatic cells, a port, such as a valve is provided in the housing for degassing, through which the gas can be extracted. In smaller cells, e.g. cylindrical cells, the gas remains in the cell. In case of pouch cells, the gas pocket is cut off and the cell is finally sealed on the open side. [29]

Innovations

Innovations in the context of degassing and final sealing of lithium-ion battery cells address in particular the reduction of process time and can influence the cell design. The elimination of a separate degassing process and the associated production station could be realized by extracting the produced gases while still forming. Immediately after formation, the gas is removed from the cell using internal pressure through a slight vacuum applied to ports or check valves. In addition to saving time, this would have a positive impact on the quality and performance of the battery cell, as the risk of plating is reduced. [30]

Another approach involves wetting the cathodes and anodes before assembly, forming the pre-wetted electrodes into an assembly, and then housing them. This process reduces the time required for the complete wetting of the electrodes and requires dedicated equipment. The gas produced during formation can escape directly, which eliminates the need for a degassing and capping station. [31]

Evaluation

Innovations in the field of degassing often influence both the production process chain and the cell design. The solutions presented have the potential to save time but require new plant technology. Since the handling of activated battery cells requires different safety requirements for fire protection in factories, it is easier to implement solutions that are based on established production process chains. The integration of degassing into the formation plant technology offers an approach with the potential for application in large-scale production.

3.4 Electrolyte Filling (2nd filling)

The electrolyte filling of large prismatic battery cells is usually conducted in two steps. The so-called 1st filling, which describes the initial electrolyte filling, is an essential part of the cell assembly (Figure 1). In practice, filling systems have several dosing units. The partially assembled cells, the electrolyte solution and the inert gas are fed to the electrolyte filling unit. The electrolyte solution is dispensed into the previously evacuated cell housing via a dosing lance [32]. Complete filling of the housing and uniform wetting of the electrodes are essential for the quality and performance of the battery cell. [33]

As a rule, large prismatic battery cells are only provisionally sealed after 1st filling or temporarily closed with a stopper if a further filling step is planned. 2nd filling is necessary to compensate for the reduction of electrolytes during the formation and to refill the cell. In the battery cell's initial charging and discharging processes, a reduction of the electrolyte can be observed due to the electrochemical activation of the cell and the formation of interphase layers such as the SEI [26]. 2nd filling only affects large-format cells, since the loss fractions of the electrolyte after formation are the highest. In addition, it is also possible to add further additives to increase the lithium-ion conductivity and stabilize the SEI layer. [22,34,35]

Innovations

An open question is the technical implementation of 2^{nd} filling. There are various concepts currently discussed in the industry, each with a different degree of innovation potential. Three noteworthy options have different advantages and disadvantages based on expert talks (Figure 2).

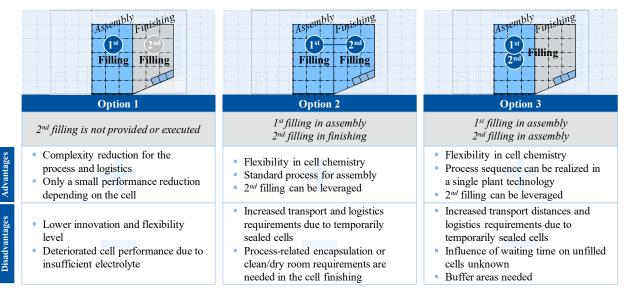


Figure 2: Advantages and disadvantages of different options for 2nd filling

Evaluation

In option 1, 2nd filling is not intended. In general, the electrolyte losses are in the low single digits [26]. Therefore, the electrochemical cell performance is expected to deteriorate only slightly (Figure 2).

In option 2, 1st filling is performed in the cell assembly and the 2nd filling is in the cell finishing (Figure 2). The advantages of second filling can be leveraged. It should be noted that prismatic battery cells must be provisionally sealed for the transfer from the assembly to the cell finishing and the initial process steps of soaking and formation. In addition, an equipment solution for the second filling is required. This results in increased space requirements and high demands on the process environment. Therefore, process-related encapsulation by mini-environments or during degassing (via a bell system) is one solution to address the dry and clean room requirements.

Another innovative idea is to bundle the two filling processes into the cell assembly ideally on the same equipment (Figure 2). The high process environment standards in the cell assembly favor the bundling of the filling stations there. However, the battery cells must be returned from cell finishing to the cell assembly and vice versa. As a result, new logistic processes and plant interfaces must be developed and defined. The increased effort favors smaller laboratory and pilot lines.

The three options promise different advantages and disadvantages (Figure 2). Depending on the constraints and desired flexibility of the planned battery factory, all three alternatives must be carefully evaluated.

3.5 Aging, End-Of-Line Testing and Grading

After formation, degassing and a potential additional 2^{nd} electrolyte filling, the battery cell is finalized. The cells are then stored for up to three weeks at room temperature in the cell finishing plant for quality assurance purposes. The cells are typically neither pressurized nor energized in trays. During storage, also called aging, the open circuit voltage (OCV) is measured at regular intervals to determine the self-discharge rate of the battery cell. [28]

The scope of the subsequent End-Of-Line (EOL) tests varies depending on the cell format, previous production process chain and manufacturer. The classical electrical measurements at the end of the production process chain are the self-discharge determined by the OCV measurements, alternating current internal resistance (ACIR) at a single frequency (typically 1 kHz), direct-current internal resistance (DCIR), and battery cell capacity calculated during formation [29]. For measurements that are typically not performed in one of the previous process steps, plant manufacturers implement separate EOL stations. These stations also include measurements of weight, dimensions, and visual inspection. Depending on the subsequent application, surface brushing and labelling or printing of the cells may also be integrated into this station. Based on the measured quality parameters determined in the EOL test, the battery cells are classified into different quality classes. [36,37]

Innovations

Innovations in the field of aging and EOL tests address the reduction of lead times and the significant space requirement. One innovative approach to reducing the significant footprint of aging by eliminating the need for long rest periods between OCV measurements is called potentiostatic aging. In this method, the cells do not rest currentless but are kept at a constant cell voltage by charging them with microcurrents. Evaluation of the charge thus induced over up to 24 hours allows the self-discharge behavior of the cells to be determined. Since the measurements are no longer performed only in specific intervals but are recorded continuously, the obtained information goes beyond the self-discharge. For instance, other defects, such as micro-shorts on the separator, can also be detected based on short-term peaks in the required charging rate. [38]

In addition to the potential future integration of electrical EOL tests into the formation procedure, there are many approaches for innovative measurement methods. By collecting extensive data and evaluating it during the aging process in adapted carrier concepts, a digital estimation of the cell quality can be made, and the aging time can be adjusted or shortened to meet the requirements of each cell. [29]

Regular EOL tests are insufficient to obtain detailed information about the condition within the cell. A possibility to gain additional data is electrochemical impedance spectroscopy (EIS). This technique can obtain information about diffusion mechanisms, charge transfer resistances and the boundary layers at the electrode surfaces. However, since EIS measurements can take up to 60 minutes per cell and require very precise measurement and power electronics, implementation in large-scale production would lead to immense additional costs. However, innovative approaches exist that make it possible to determine the entire frequency spectrum of an EIS measurement within a few seconds by pulse analysis, thereby drastically accelerating the process without significantly degrading the information value. [39,40,41]

Evaluation

Metrological innovations in the field of aging or EOL testing, which pursue the purpose of shortening aging, have the potential for significant cost and space savings in production operations. In principle, potentiostatic aging can be integrated, but this requires adjustments to the cell finishing plant concept. The integration of electrical EOL tests into the formation procedure as well as AI-based quality prediction can be implemented with existing equipment concepts and therefore offer feasibility in volume production. By integrating pulsed impedance measurements, the technology can be implemented in large-scale production that is usually limited to the laboratory scale.

3.6 Sorting and Packaging

The cells are sorted into several classes according to the specifications ("grades") and filled into packaging units on holding structures in an automated manner for high-volume production. For safety-critical cells, packaging in safety transport containers is typically required. These packaging units are dispensed onto a conveyor belt or a facility for temporary storage with a specified capacity. The packaging units are then placed on a pallet. The capacity of the intermediate storage facility must be matched to the production campaigns so that the transport of the batteries to the storage facility can take place in the targeted schedule. [28,42]

Innovations

The automation degree of the packaging unit is a central adjusting lever for throughput and innovation. In large-scale production with high throughputs, fully automated AGVs (Automated Guided Vehicles) and multi-axis industrial robots are used. These robotic arms allow maximum utilization of the workspace and flexibility. [43]

Additionally, the packaging must meet a variety of requirements for lithium-ion battery cells (UN certifications). In addition to ensuring the structural integrity of the cells, increased safety requirements must be considered. The fire hazard of batteries can be mitigated by the packaging. Modern packaging solutions provide improved flame retardance and arrestment, thermal management, pressure management, blast containment, and gas and smoke filtration. Furthermore, re-engineering allows for a reduction in product complexity through new material solutions while increasing the intrinsic safety of the packaging. [44, 45]

Evaluation

The degree of packaging automation is essential for large-scale production to achieve cost and time reductions. However, the leverage at the end of the process chain is limited since the process cycle times have lower reduction potential in comparison to the processes in electrode production, cell assembly or cell finishing. Furthermore, automation solutions in the packaging process are characterized by cross-industry adaptability and applicability in scaled productions. The same applies to modern packaging solutions that can be used universally in production (e.g. storage and transport) and sustainable packaging materials.

4. Results and Discussion

The innovations presented along the process chain in cell finishing were assessed regarding their potential and applicability in large-scale production. The assessment is based on insights from leading industry experts and publications in the field of cell finishing. The potential refers to the possible time and cost savings achieved with the innovations. Furthermore, the applicability of the innovations remains significant, as this determines whether the innovations can be used in large-scale production or if increased implementation effort is necessary. In addition, the scaling level of the concept must be technically achievable.

It can be stated that the process innovations in cell finishing mainly focus on the bottleneck processes with the longest cycle times and largest investment requirements. The soaking, formation, and aging can be identified as such processes. There, experience and an understanding of the electrochemical principles are crucial. New process protocols and existing equipment technology will enable time reductions and potentially reduce energy costs.

In addition, quality-enhancing measures such as 2^{nd} filling are becoming increasingly important. Here, the focus shifts depending on the objective of cell production. The innovations presented must be adapted to the logistics and environmental constraints of the factory concept and the desired flexibility of production.

It can be expected that these innovations will be associated with increased costs in the initial phase and will typically move in first where high-quality and more costly battery cells are produced. That's where the plant technology's increased costs and integration efforts are profitable at first. This also means that in large-scale production, the trade-off between low cost (\$/kWh) and quality results in a much longer time horizon before new process innovations can be introduced.

5. Conclusion and Outlook

New process innovations are supporting the development of so-called Gigafactories in Europe [46]. As well as improving product quality, these technologies could reduce costs and accelerate production times significantly. At the same time, sustainability through energy savings and thus an improvement in energy consumption through process innovations and further developments in plant technology is increasingly important. Research and industry efforts make it possible to exploit this potential. Batteries with higher requirements will be the first to benefit from the upcoming innovations due to the current cost pressure in battery cell production. Trickle-down effects into more price-sensitive cell production are expected mid to long-term. These activities and developments will be complemented by research factories, which will enable European machine and plant manufacturers to bring their process innovations into series production sooner.

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