Design of an Automated System to Accelerate the Electrolyte Distribution in Lithium-Ion Batteries

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***Abstract*—During the last years lithium-ion batteries have emerged as the power source of choice for the electric vehicle market. One important step of the lithium-ion battery process chain is electrolyte filling. The electrolyte filling process constitutes the interface between cell assembly and formation and is a quality critical and also time consuming process step. To avoid limitations in battery performance, a homogeneous electrolyte distribution is necessary. Therefore, especially large cells are stored for hours to reduce their wetting time. In order to reduce this wetting time, an automated system to accelerate the electrolyte distribution in lithium-ion batteries by press rolling was designed. For the technical design concept, the degree of kinematic coupling between roller and the drive system for cell transport was determined. Subsequently, an exemplary motion profile and resulting loads due to the rolling process were derived in order to design the drive system. To quantify the influence of press rolling on electrolyte distribution a transparent test cells had been developed for visualization. Finally, a procedure for evaluating the optical data of the macroscopic electrolyte distribution was deducted.**

***Index Terms*— electrolyte filling, lithium ion battery, press rolling, wetting**

I. Introduction

In the last years, lithium-ion batteries have emerged as the power source of choice for the electric vehicle market [1]. The process chain of a lithium-ion battery production can be divided into three main sections. These sections contain electrode production, cell assembly and electrochemical characterization [2][3][4]. At the interface of cell assembly and electrochemical characterization, the electrolyte filling process takes place. This article focuses on electrolyte filling, because the procedure is quality-critical and also time-consuming [5][6].

The quality of a battery is influenced by the macroscopic electrolyte distribution between porous battery sheets (electrodes, separator) and the soaking behaviour of these porous structures on microscopic scale. Less wetted areas on both scales influence the battery performance by increasing the electrolyte resistance. As a result, the high current charging and discharging ability decreases [7]. In order to avoid these limitations in battery performance, a homogenous electrolyte distribution is necessary. Therefore, especially large cells are stored for hours [8].

This duration is known as wetting or soaking time. Wetting of a stack or electrode-separator-compound (ESV) is driven by capillary forces. To accelerate the macroscopic filling and microscopic soaking processes cells are filled under low pressure conditions in a vacuum chamber [9]. Also, cycles of alternating pressure are applied to foster these infiltration processes [11] [12] [13].

Whether the stack inside the cell is sufficiently wetted can be evaluated after filling and wetting by determining the electrochemical battery performance. In [14], this method was applied to determine the influence of two different pressure profiles on discharge capacity. The authors of [15] examined alternating courses of pressure applied during and after filling. Differences in electrolyte distribution were visualized by neutron radiography. Approaches to accelerate the electrolyte infiltration by means of a mechanical device are described in patents [9][16] and industrial approaches [17]. Effects on electrolyte distribution or battery performance have not been evaluated systematically.

In summary a few approaches are known, which demonstrate a positive effect of pressure on optical electrolyte distribution and battery performance. Despite this potential, research activities in this field are rarely observed. Research activities are also necessary because the automotive industry tends to produce thicker and larger cells to increase the energy density of battery cells. The wetting time and also costs increase with these upscaling tendencies [18].

Therefore, this paper intends to design an automated system to accelerate the electrolyte distribution in large scaled lithium-ion batteries. This system comprises of a linear axis drive, for transferring the battery cell, and a combined screw drive, which provides vertical rolling force onto the cell. The roller is intended to apply a line load to the cell in order to induce cell-internal shear forces. It can either be actively controlled or implemented as a passive element which is only moved due to the linear motion of the battery cell. In the following section, the technical design concept for the rolling system is concretized and the components for the linear axis drive are configured according to the requirements. Subsequently, the authors propose an optical characterization method to visualize the influence of press rolling on electrolyte distribution. In addition, an

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evaluation process for the optical data of the macroscopic electrolyte distribution is deducted.

II. Technical Design Concept to Accelerate the Electrolyte Distribution

For the technical design concept, at a first step, the degree of coupling between roller and linear axis central was determined and three different coupling concepts were developed. For an adequate linear axis drive configuration, an exemplary motion profile for the rolling process was developed at a second step. In addition, the occurring roller forces are significant for the configuration of the horizontal linear bearings which provide guidance for the battery cell carrier. As a consequence, the third step contains implementing four carriage bearings for this purpose. Three different load cases were investigated considering force transmission between roller and cell carrier.

A. Requirements

Multiple requirements to be met by the rolling system have been identified. For example, the rolling system is intended to process cells of three different sizes BLB1, BLB2 and BLB 3 (80x110mm, 140x190mm and 240x340mm). In preliminary tests, the maximum applicable rolling force was examined. By use of a modified uniaxial pressure test, multiple cells were stressed until failure. As a result, a maximum rolling force of 15 kN was defined.

Geometrical requirements depend on the given laboratory environment. Moreover, it is intended to implement adequate measurement technology in order to provide in-line process monitoring. In Table I, the most significant requirements are listed.

TABLE I. Requirements

Category	Description	Value
Geometry	Dimensions	800 x 1500 mm
Kinematics	Vertical roller travel range	100 to 400 mm
	Linear axis travel range	800 to 1200 mm
	Possible roller force	15 kN
Sensors	Roller force detection	15 kN
General	Overall weight	80 – 150 kg

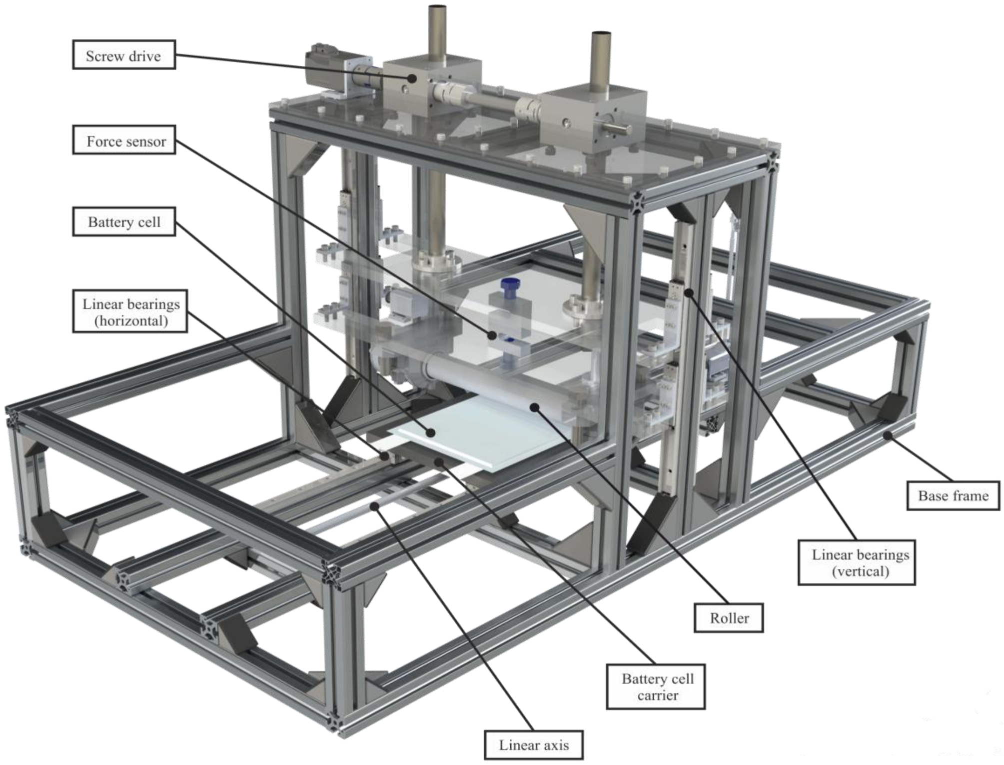

Figure 1. Design concept of a press rolling device

All identified requirements were used to develop the proposed design concept. The following sections describe detailed kinematic and configuration considerations for a well-working rolling system.

B. Coupling of Roller and Linear Axis

For the technical design concept, the central issue is to determine the degree of coupling between roller and linear axis. In particular, three different coupling concepts were developed (Fig. 2). Whereas roller and linear axis can be coupled both in a merely mechanical (a) and a feedback controlled manner (b), the third concept does not include any coupling mechanism (c).

Since feedback controlled coupling allows for free choice of drive parameters, mechanical coupling only provides a fixed roller speed which is dependent on the linear axis' speed due to a constant gear ratio. Conversely, despite the lack of controllability, a non-coupled roller configuration does not require further equipment, which reduces both cost and likelihood of errors.

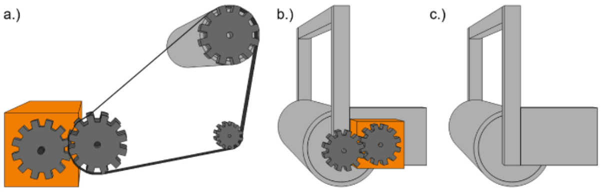

Figure 2. Coupling concepts

However, it has been planned to implement the most cost efficient concept (c) at a first step and evaluate the results. The addition of a roller drive system is still possible, if needed in the future.

On the basis of the above-mentioned considerations, firstly, a schematic conceptual design was elaborated. At a second step, the rolling system design was concretized by selection of specific products, on the basis of the previously mentioned requirements. The final design is shown in figure 1. Screw drives were selected for the vertical roller motion, providing self-locking capabilities and high forces. In this context, it is further intended to implement a steel roller from solid material in order to provide sufficient force transmission to the cell specimen.

For system automation, it is planned to implement a programmable logic controller (PLC) system. In a multi-criteria analysis – considering criteria such as machine-to-machine (M2M) communication capabilities, extensibility and cost – PLC systems of multiple suppliers were compared against each other. The analysis resulted in the selection of the compact PLC system CX9020 by Beckhoff, since it offers the richest M2M capabilities as well as simple extensibility and high computational power. Moreover, this PLC system runs on a common Windows computer which offers the opportunity to implement third-party software such as, for example, code generated in MATLAB. Control devices and power supply are not shown in Fig. 1 for a cleaner view on the design.

C. Configuration of Linear Axis Drive

The linear axis drive provides the cell carrier motion and is therefore exposed to the same load that is applied to the battery cell. This leads to internal friction forces. In addition, certain inertial forces occur due to de- and acceleration. Therefore, the authors propose an exemplary motion profile for the rolling process (see Fig. 3), which serves as a basis for calculating the minimum required motor torque for the linear axis drive. The profile comprises the following stages:

- **$t_0 – t_1$:** Equip the cell carrier with battery cell and accelerate up to 25 mm/s.
- **$t_1 – t_2$:** Move with constant velocity to approach start position of rolling process.
- **$t_2 – t_3$:** Decelerate for reaching the start position.
- **$t_3 – t_4$:** Apply roller force onto the battery cell and accelerate carrier to target speed 5 mm/s.
- **$t_4 – t_5$:** Rolling process at constant carrier speed.
- **$t_5 – t_6$:** Accelerate up to transport speed (25 mm/s).
- **$t_6 – t_7$:** Decelerate until stop.

From the maximum applied rolling force (15 kN) onto the cell carrier results a static friction force of 46 N. Thus, the linear axis drive must offer linear forces above this friction force at the desired carrier acceleration. This leads to a minimum torque of 200 Nmm in order to reach 25 mm/s at the desired acceleration.

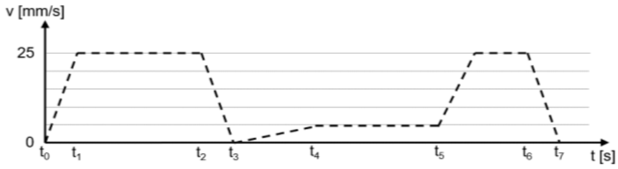

Figure 3. Exemplary motion profile for linear axis drive

For the screw drive, an overall torque of 3.5 Nm has been computed which is required to provide the maximum rolling force of 15 kN.

D. Configuration of Carriage Bearings

The applied rolling forces are also significant for the configuration of the horizontal linear bearings which provide guidance for the battery cell carrier. Four carriage bearings were implemented for this purpose. Three different load cases are investigated considering force transmission between roller and cell carrier. In general, the load working on each of four carriage bearings can be computed by use of (1), whereas F_R stands for the applied roller force, l_1 describes the load displacement in y and w_1 the displacement in x.

$$L = \frac{F_R}{4} \pm \frac{F_R \cdot l_1}{2 \cdot l_0} \pm \frac{F_R \cdot w_1}{2 \cdot w_0} \quad (1)$$

Fig. 4 depicts these three load cases for line loads on the cell carrier. The four carriage bearings are labelled as C1, C2, C3 and C4. Due to symmetry, loads on C1 are always equal to those on C2, which is analogue for C3 and C4. Case 1 describes the roller applying a line load onto the frontal or rear edge of the cell carrier. This results in $L_{1,2}$=7.03 kN for the frontal bearings and a significantly lower load $L_{3,4}$=0.47 kN for the rear bearings. Thus, both frontal bearings absorb almost the entire applied roller force. Whereas in Case 2, which schedules a load displacement of a fourth of the total length l_0, the load distribution provides $L_{1,2}$=5.16 kN for the frontal bearings and $L_{3,4}$=2.34 kN for the rear bearings, the loads are equally distributed in Case 3 (the line load is placed in the exact center of the carrier). Since Case 1 is clearly the most critical load case, it has been decided to select carriage bearings KWVE-20 B by Schaeffler INA, which offer a dynamic load rating of 13.1 kN as well as a static load rating of 27 kN.

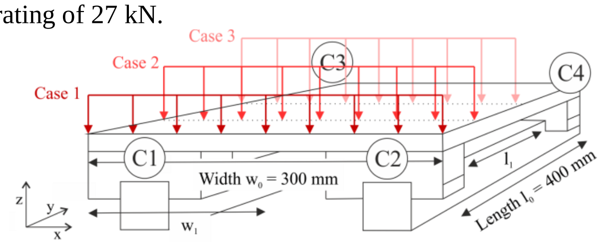

Figure 4. Line load cases on carriage bearings

III. Analyzing the Effect of Press Rolling on Electrolyte Distribution

After designing a mechanical device to accelerate the electrolyte infiltration, it ́s also important to quantify the effect of press rolling on electrolyte distribution inside the battery cell. A method was developed to allow an optical ex-situ characterisation of the electrolyte distribution. Therefore, transparent cells are assembled and filled with electrolyte. Then an evaluation process for the optical characterization of the macroscopic electrolyte distribution was deducted using MATLAB and its image processing toolbox.

A. Preparation of a Transparent Lithium-Ion Battery

To enable the visualization of the electrolyte distribution transparent cells have been built (Fig. 5).

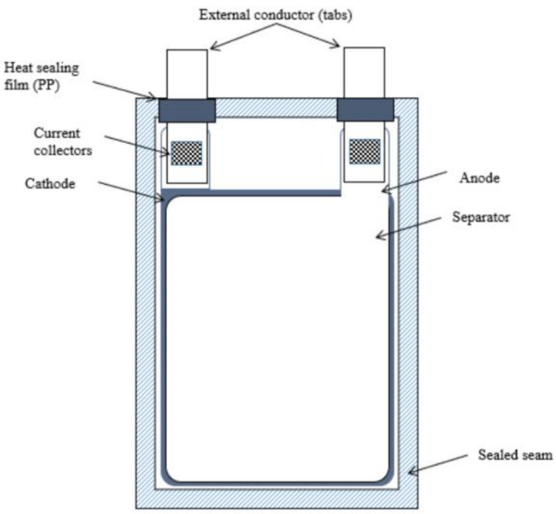

Figure 5. Design of a transparent test cell

The main components of a common LIB and as a consequence of the transparent cells are electrodes, separator and electrolyte which in case of a so called pouch cell are housed in a polymer laminated aluminium foil. Before housing, a defined number of electrodes and separator sheets are stacked or winded to a compound and contacted to achieve the desired battery performance.

Electrodes are usually composed of a current collector substrate coated with an electrochemically active porous composite. For the transparent cells a cathode with $LiNi_{0.33}Co_{0.33}Mn_{0.33}O_2$ as active material coated on 10 µm copper current collector and a graphite (SMG-A5) anode on a 20 µm aluminium foil was used. Electrodes are separated by a commercial Al2O3 based separator (SEPARION® S240P20, Litarion GmbH). This electrically insulating porous ceramic is characterised with a thickness of 21 µm, a porosity of 48 % and a gurley number of 20 s. Cathodes are tailored to BLB2 standard with a shape of 105 mm*145 mm, anodes to 110 mm*150 mm and separator to 370 mm*156 mm. The cells were manufactured in the drying room of the Battery LabFactory Braunschweig. The electrode-separator compound (ESC) was z-folded. After ESC assembling it ́s common practice to enhouse the ESC in a bag containing of two sheets of laminated aluminium foil. Therefore, commercial available pouch foil is used and sealed on three sides. As a consequence, the bag is still open so that the ESV and also the electrolyte can be filled inside. For the transparent cells only one pouch foil sheet - instead of two - was used. The second one was substituted by a polyethylene foil (PE). The PE foil was chosen because of its high transparency, chemical electrolyte resistance and good sealing behaviour. After sealing the pouch foil one the one side and the transparent PE-foil on the other, the ESV was put inside. Then the cell was filled with V = 1 ml electrolyte. Therefore, commercial electrolyte LP57 (1mol LiPF6, EC/EMC with mass ratio 3:7) from BASF was used. The filling procedure was done in one step under low pressure conditions. Finally, the transparent cell was closed by sealing the last of the four sides.

B. Deduction of a Quantification Process

After designing and manufacturing a transparent cell an evaluation process for the optical characterization of the macroscopic electrolyte distribution in a transparent lithium-ion batteries was deducted. The filled an d sealed cell was fixed in a workpiece carrier. A camera system was placed in front of the transparent cell and the changes inside the cell were recorded for a duration of two minutes. As shown in Figure 6 it was possible to visualize dry or unsoaked areas on the cell surface. It’s also shown that these areas decrease over time, which indicates an increase in electrolyte distribution and as a result the wetting degree.

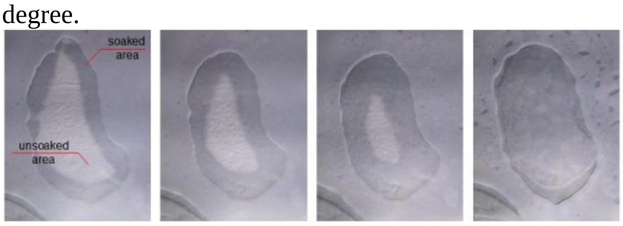

Figure 6. Increase of wetted area over time inside transparent battery cell.

To quantify the electrolyte distribution, the recorded video was analysed with MATLAB and its image processing toolbox. In a first step the coloured was transformed into a greyscale image. Second a histogram is given to the grayscale image, indicating the number of times each grey level occurs in the image. The contrast of the images was enhanced by spreading out the achieved histogram [10]. That’s necessary to separate completely soaked from dry or less wetted areas in the cell. As a result, using this contrast enhancement tool enables the visualisation of the macroscopic electrolyte distribution. Besides visualizing the electrolyte distribution also gas inclusion could be recognised. Gas inclusions were induced by applying low pressure during filling process. These effects influence the battery performance in a negative way, because they cause less wetted and hence inactive areas.

To contain more information about e.g. size of dry areas, in a third step an edge finding algorithm was used. This algorithm enables to create edge images by using a Sobel filter and the threshold was determined manually for each image. As a result, gas inclusions and dry areas could be separated from the used ceramic separator more clearly. Finally, the amount and average size of gas inclusions and the electrolyte distribution in Lithium Ion Batteries was determined.

In summary, a method was developed to allow an optical ex-situ characterisation of the electrolyte distribution. Therefore, transparent cells were assembled and filled with electrolyte. Then an evaluation process for the optical characterization of the macroscopic electrolyte distribution was deducted. Therefore, MATLAB and its image processing toolbox was used and histograms, edge detection and thresholding techniques were applied.

IV. Conclusion

An automated system to accelerate the electrolyte distribution in Lithium Ion Batteries was designed. For the technical design concept, the degree of coupling between roller and linear axis central was determined first and three different coupling concepts were developed. For an adequate linear axis drive configuration, an exemplary motion profile for the rolling process has been developed in the second step. In addition, the occurring roller forces are significant for the configuration of the horizontal linear bearings which provide guidance for the battery cell carrier.

As a consequence, the third step contains implementing four carriage bearings for this purpose. Three different load cases were investigated considering force transmission between roller and cell carrier. Finally, an optical characterization method was developed to visualize the influence of press rolling on electrolyte distribution. Therefore, transparent test cells had been successfully developed. Additionally, an evaluation process for the optical data of the macroscopic electrolyte distribution was deducted. Therefore, histograms, edge detection and thresholding techniques were applied. The effect of time on the amount and average size of gas inclusions and the electrolyte distribution in Lithium Ion Batteries could be visualized and evaluated. In conclusion first steps are done to evaluate the effect of press rolling on electrolyte distribution of large scaled lithium- ion batteries.

Finally, this study is a necessary work to reduce the wetting time of large scaled LIB systematically and to avoid limitations in battery performance by an inhomogeneous electrolyte distribution.

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