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### Concepts for the Reduction of Longitudinal Wrinkles During Calendering of Battery Electrodes

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#### Abstract

Many potential customers are currently still discouraged from buying an electric car because the prices are significantly higher than their expectations. Important cost drivers are the high amount of energy required to produce the battery cell as well as high material costs. These costs can be reduced by minimizing scrap in manufacturing, because less energy and material is wasted. The calendering process step is a rolling process in which the coating of an electrode is compacted. Among many other positive effects, calendering is a necessary process step to increase the volumetric energy density. However, calendering can also generate defects in the electrodes caused by the high mechanical stresses due to high line loads during compaction. One such defect is the formation of longitudinal wrinkles that form at deflection rollers. They lead to scrap and, in the worst case, to web tears that cause process interruptions. This paper presents two material-independent concepts to reduce longitudinal wrinkling during calendering. One contributing factor is the deformation of the electrode during calendering. Therefore, the aim is to minimize the deflection of the electrode out of the plane, so that the web can hit the deflection roller straighter and longitudinal wrinkling is reduced. The first concept involves the implementation of additional rollers and the second concept realizes a lateral stretching movement. This paper also shows the implementation of prototypes, first results and recommendations for an optimization. Finally, further concepts can be developed on the basis of these findings in order to reduce scrap and thus energy wastage and production costs. Consequently, a more sustainable and resilient electrode manufacturing will be enabled. The material-independent approaches will also allow for the implementation in the electrode production of new and sustainable battery concepts.

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#### 1. Introduction

Compared to an internal combustion engine (ICE) vehicle, the cost of a battery electric vehicle (BEV) is still very high, which discourages many potential customers from buying a BEV [1]. Using a lithium manganese oxide (LMO) graphite battery pack, Yuan et al. showed that 47 % of the total energy required during production is used for coating and drying. The energy consumption of the drying room is also high at 29 %. [2] Using the example of a lithium nickel manganese cobalt oxide (NMC) graphite cell, it was shown that over 70 % of the total costs are referred to the materials, while the energy costs are comparatively low at 3.1 %. [3] At the same time, the scrap rates of the individual process steps are estimated to be at up to 5 %. However, production can only become efficient when the scrap rate falls below 1 %, while experience still needs to be gathered at many production sites. [4] In addition to operator experience, an in-depth understanding of the process is also necessary to reduce the scrap rate. This work therefore focuses on the reduction of scrap during the calendering process. When calendering, the electrode is compacted in a rolling process to increase the volumetric energy density of the cell [5]. This requires high line loads, which cause defects such as waves, foil embossing and many more [6].

The main objective of this work is the development of concepts to reduce the defect longitudinal wrinkle, which has

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already been introduced and discussed in [7,8]. Longitudinal wrinkles are plastic deformations in the running direction of the electrode that can lead to web tears and are also known from other web processing industries [9,10]. In this work, only longitudinal wrinkles in the uncoated part of the substrate are considered.

There are already a number of solutions for reducing longitudinal wrinkling in the processing of thin webs of paper, plastic or metal, for instance. A widely used concept is the socalled spreader roller, which can be realized in various shapes. They are designed to introduce continuous forces into the web that act transverse to the running direction and thus induce stretching of the web [11]. For example, there are rollers with a curved axle, as presented in [12,13]. There are also many variants of straight rollers with spiral profiling, such as in [11,14]. Furthermore, so called crown rollers are used before winding web material. The diameter of these rollers decreases from the center outwards. [15] In [16] the application for winding electrode webs is demonstrated. [17] and [18] show a deflection roller for guiding electrodes with an uncoated substrate edge, which only touches the coated part of the electrode. A list of common spreading methods is also given in [19]. However, no scientific reports on the use of these solutions in battery production and especially during calendering are known at this time. In the following, the conception and validation of two potential concepts for the reduction of longitudinal wrinkling in battery electrodes are presented.

#### 2. Concept Development

The development of the two concepts is based on VDI 2221 [20]. First, the requirements for the system are identified. Then the structure of functions and principle solutions are created. Using a morphological box, the best partial solutions for the two concepts are selected and worked out for implementation as a prototype.

#### 2.1. Requirements

The system under consideration is the GKL 500 MS calender (Saueressig Group) already presented in [7,8]. Longitudinal wrinkles form on the first deflection roller after calendering [7]. Under the given requirements (high density, high web tension), the formation of longitudinal wrinkles cannot be prevented by changing the process parameters. Therefore, an additional device must be integrated that affects the process so that longitudinal wrinkling is prevented or at least reduced under the required process conditions. One further advantage is that an additional device is independent of the electrode material. The installation space results from the structure of the tension unit of the calender. The deflection rollers are firmly mounted in a steel frame. As shown in [8], the electrode is guided straight out of the calender up to the first deflection roller and is then deflected in a S-shape. As shown in [8], the longitudinal wrinkles are formed as soon as they hit the deflection roller, with the formation zone starting a few centimeters earlier. The additional device must therefore be fitted as close as possible to the deflection roller. Furthermore,

the course of the web is not to be changed. Since electrodes with different dimensions should be processable in the future, the device should be as flexible as possible in its position. As this is initially a prototype, which is intended to gain knowledge in order to derive measures for improvement, the design should be kept simple, whereby the decisive components should be easy to assemble and disassemble. A certain degree of adaptability should also be ensured. The complexity of the system's control should be low to allow fast commissioning.

The first main function is to apply a force  $F_{str}$  that is sufficient to stretch the web in the x-direction. As shown in Fig. 1, this stretching force should be applied on the uncoated edge of the substrate in order to avoid damaging the coating. The surface pressure  $F_{SP}$  indicated in Fig. 1 must be overcome so that the electrode can be fully pressed against the deflection roller. In the first step, only electrodes with the same uncoated conductor edge on both sides are considered. The structure should therefore also be symmetrical to ensure uniform force distribution and to avoid changing the alignment of the web.

The second main function is to guide the electrode so that it hits the deflection roller as flat as possible.



Fig. 1 Schematic of a longitudinal wrinkle in an electrode at the deflection roller according to [8] with an additional stretching force  $F_{str}$ 

#### 2.2. Functional Structure and Principal Solutions

The functions are analyzed using the function analysis system technique (FAST) according to [21]. The device must supply the mechanical energy and transfer it to the electrode in order to apply the stretching force and guide the electrode. In general, the force can be transmitted by means of geometric locking or frictional connection, whereby the force can be continuous or discontinuous. As can be seen from section 1, rollers are predominantly used for continuous operation in web processing. The mechanical energy can be provided by the rotation of the roller or by an external drive or braking torque. Discontinuous force transmission is defined here as an iterative application of force with a constant duration. This means that there are force-free time intervals. The morphological box of the target system, shown as Table 1, summarizes the potential partial solutions for the functions. The roller concept is shaded in light gray and written in italics, while the finger gripper concept is shaded in dark gray and written in italics with white font.

Table 1. Morphological box with the functions and partial solutions

	Function		Partial solution	n
		1	2	3
А	Apply force in	D - 11	Spring	Electro-magnetic
	x-direction	Koller		actuator
В	Guide	Rolling	Sliding	
	electrode	contact	contact	-
С	Drive	Passive	Electric drive	-
D	Force	Divert	Cam	Tintere
	transmission	Direct	mechanism	Linkage
Е	Contact surface	Rigid	Soft, non-slip	Soft, gliding

#### 2.3. Concept 1: Additional Rollers

The morphological box in Table 1 shows the partial solutions for the roller concept, which is presented in Fig. 2.



Fig. 2 CAD model of the entire roller concept

Two rollers are used, which are pressed symmetrically from above onto the coating free substrate edge. In this way, the two main functions are realized. Both rollers are fastened in a holding fixture, which can be seen in Fig. 3a). The rollers are mounted in a carrier with adjustable angle and are screwed into a rail with a slotted hole so that they can be positioned in the direction of travel. This allows the distance to the deflection roller to be adjusted. The strength of the contact pressure of the rollers on the substrate is defined by the distance in the zdirection, which is set by the number of spacers used.



Fig. 3 a) Roller holder, b) Roller geometries: cylindrical (CY), barrel-shaped (B), conical (CO), cylindrical with spiral profile (S)

The roller holders can be moved individually in the xdirection on a support rail, which is realized as an aluminum profile as shown in Fig. 2. The ability to position them in the xdirection allows a flexible adjustment to format changes. The carrier is firmly connected to the frame of the tension unit by screw connections via an anti-twist carrier plate. All components can be assembled and disassembled, making it possible to replace the components.

Four geometries are selected for the rollers, which are shown in Fig. 3b). Rounded edges on all rollers prevent damage to the electrode. The first shape is the cylindrical shape (CY), which corresponds to a simple deflection roller. A barrel shape (B), which is based on the convex spreader rollers mentioned in section 1 and a conical shape (CO) are selected as further geometries. The diameter decreases towards the outer edge of the web, which is intended to induce an outward force. Finally, a spiral profile (S) is designed into the simple cylindrical shape. The rollers are made of thermoplastic polyurethane (TPU) with a 95 Shore A hardness (Ultimaker B.V.) using 3D printing.

# 2.4. Concept 2: Finger Grippers for Stretching in the Transverse Direction

The partial solutions for the finger gripper concept are taken from the morphological box in Table 1. Fig. 4 shows the CAD model of the entire concept.



Fig. 4 CAD model of the entire finger gripper concept

The pair of finger grippers is positioned symmetrically and clamps the electrode on the uncoated substrate, while pulling it outwards in opposite directions. For a uniform stretching process, the fingers should move simultaneously and with identical frequency and clamping force.

Fig. 5 shows a detailed sketch of the finger gripper.



Fig. 5 Sketch of the finger gripper system

The finger gripper consists of two fingers, which are mounted on the so-called finger plate and are connected by gears. The closing force of the fingers is generated by a springdamper system. As shown in Fig. 6, the opening and closing of the finger pair is realized by a lever mechanism. The lever is rotatably mounted on a fixed part, which does not move forward with the plunger. During the forward movement, the rotational movement is limited by a stopper so that it presses the fingers open during the forward movement (2). The stopper is also mounted on the fixed part. The finger gripper moves forward to the reversal point at which the contact between finger and lever breaks off and the fingers close (3). The closing mechanism is slowed down by a damper to ensure that the finger gripper does not move forward when closed, as otherwise web tears may occur. In Fig. 6, the damper is not displayed for simplicity. In the reverse movement, the lever can rotate freely and move around the end of the finger back to the contact-free starting position (5). The finger gripper remains closed due to the spring.



Fig. 6 Motion sequence of the finger gripper, shown for one finger only for clarity

An external drive is required for the periodic movement of the finger gripper. It is designed as an electrically driven cam gear in the form of a cam cylinder for each finger gripper. The rotation of the motor is transmitted to the cam cylinder by a toothed belt with a transmission ratio of i = 1. The rotary movement of the grooved cam cylinder is translated into a cyclical linear movement of the plunger by the groove milled into the lateral surface. The plunger and grooved cam are coupled by a roller. The plunger is screwed onto the finger plate, which in turn is mounted on the slide of the slide guide. The motor and the cam cylinder are fixed to a mounting plate that is bolted to the frame of the tension unit. Both motors are controlled using a joint Arduino Mega 2560 Rev3 microcontroller (Arduino S.r.l.).

#### 3. Validation

The following validation tests evaluate the functionality in the application with calendered electrodes.

#### 3.1. Electrode Materials and Experimental Parameters

The NMC811 electrode with the 30 mm wide symmetric uncoated substrate already examined in [8] is used for the validation. The maximum requirement of highest density  $(\rho_{NMC} = 3.3 \ g \cdot cm^{-3})$  and highest web tension  $(F_w = 110 \ N)$ is selected from the process window of density, web tension and temperature examined in [8] for testing both prototypes. The roller temperature is selected as  $T_r = 30 \ ^{\circ}C$ .

#### 3.2. Concept 1: Additional Rollers

Using the spacers, the distance between the rollers and the electrode is minimized so that a slight contact pressure is created as displayed in Fig. 7a). The rollers are first aligned

parallel to the deflection roller ( $\alpha = 0^{\circ}$ ) and then examined at an angle of  $\alpha = 10^{\circ}$ , as shown in Fig. 7b).



Fig. 7 Installation situation of the rollers in the experiment

The functionality of the roller types depending on the set angle  $\alpha$  is shown in Table 2. At an angle of  $\alpha = 0^{\circ}$ , none of the roller concepts were able to prevent the formation of longitudinal wrinkles. The outward driving force was also too low for the cylinder with the spiral profile. Changing the angle to  $\alpha = 10^{\circ}$  could not bring about any improvement for the cylindrical and barrel-shaped rollers. However, the conical rollers and the spiral profiled rollers prevented longitudinal wrinkling. The conical rollers behave in a similar way to the convex spreader rollers already discussed. They stretch the material to the side with the narrower diameter. This is also the reason why the barrel-shaped rollers do not prevent longitudinal wrinkling. These push the electrode in both directions, thus also towards the center of the electrode. With straight alignment, the force in the x-direction is not sufficiently strong. The angled alignment, on the other hand, increases the force in the x-direction. Similarly, the inclined alignment increases the proportion of the force in the xdirection caused by the spirals.

Table 2 Overview of the functionality of the various rollers at different angles

Angle $\alpha$ / °	Cylindrical	Barrel	Conical	Spiral
0°	No	No	No	No
10°	No	No	Yes	Yes

The surface of the calendered electrode is scanned with the LJ-X8900 line laser sensor (KEYENCE DEUTSCHLAND GmbH) to assess the guidance of the electrode. The laser is fixed above the electrode at a distance of 320 mm from the axis of the deflection roller, while the web moves during the measurement. The scanning frequency is 50 Hz. Fig. 8 shows a comparison of the averaged cross-sections of the NMC811 electrode obtained from the laser data with and without the additional rollers.



Fig. 8 Averaged cross-section in x-direction through the laser data of the calendered NMC811 electrode with and without additional rollers

The 30 mm wide uncoated substrate edges of the electrodes behave differently. The edges of the web that are guided by the rollers are significantly flatter and tend to be inclined upwards compared to the edges of the unguided electrode. This leads to a straighter hitting of the substrate on the deflection roller and inhibits the formation of longitudinal wrinkles.

Fig. 9 shows photos of the calendered NMC811 electrodes using the conical and spiral profiled rollers with  $\alpha = 10^{\circ}$ . There are no longitudinal wrinkles in the uncoated area. However, due to their rough surface, the rollers leave an imprint on the substrate, which needs to be optimized in future developments. The defect pattern foil embossing is also clearly visible, but this is not the aim of the optimization and is therefore neglected at this point.



Fig. 9 Longitudinal-wrinkle-free calendered electrodes: a) conical rollers, b) spirally profiled roller

# *3.3. Concept 2: Finger Grippers for Stretching in the Transverse Direction*

Table 3 shows the variations investigated using the finger grippers. First, two different rough needle felts made of polyethersulfone (PES) were initially tested in order to minimize the risk of web tears. The roughness can only be compared subjectively, as no information is available from the manufacturer. Furthermore, two types of rubber were examined. The first material is a structured anti-slip rubber slider. The second material is an SBR rubber strip. For both rubbers no further material details were found either.

Table 3 Experiment design to validate the finger gripper concept

No.	Gripper surface	Spring force / N	Motor speed / rpm
F1	Smooth felt	4.67	35
F2	Rough felt	4.67	35
F3	Structured rubber	8.52	35
F4	Structured rubber	8.52	70
F5	Smooth rubber	8.52	35
F6	Smooth rubber	12.37	35

Fig. 10 shows the material selection. The materials are cut to size and glued to the finger tips as a contact surface, which is 40 mm<sup>2</sup> per finger. The grippers have two springs on both sides that define the clamping force. The influence of the motor speed is also investigated. The damper is not varied as the closing time should not be changed.



Fig. 10 Contact surface material: a) smooth felt, b) rough felt, c) structured rubber, d) smooth rubber

The longitudinal wrinkling showed no measurable difference for both felts. It is therefore necessary to further increase the friction between the gripper and uncoated substrate. This is achieved by using the rubber sliders and increasing the clamping force by increasing the spring force. Fig. 11 shows the heights of the longitudinal wrinkles for the different variations using the rubber sliders. The data was recorded with the laser in the same way as already presented in [7]. The evaluation could not be performed with the MATLAB script from [7], as the longitudinal wrinkles form too close to the coating edge. For this reason, it is not possible to specify the distance. Furthermore, the width cannot be measured reliably either, as the side facing the coating edge blurs with the coating edge. The height was evaluated using the LJ-X Observer software (KEYENCE DEUTSCHLAND GmbH).

Fig. 11 also shows the corresponding test no. #6 from [8] (Ref.) without the intervention of the finger grippers as a comparison. The initial visual impression that the longitudinal wrinkle formation is influenced is confirmed. The height of the longitudinal wrinkles decreases when the finger grippers intervene. At F6 the reduction is about 48.4 % compared to the reference. From preliminary tests, in which the web was manually stretched to the side and guided, it is known that a lateral force can prevent the formation of longitudinal wrinkles. Therefore, it is assumed, that the applied stretching force is too low to flatten the electrode sufficiently and therefore prevent longitudinal wrinkling. The increase in the stretching force due to the measures taken also appears to be minimal, so that no clear influence can be identified. However, it seems like the smooth rubber with a larger contact surface compared to the structured rubber and a higher spring force is the best solution. Nevertheless, the system must be further optimized.



Fig. 11 Change in the height of the longitudinal wrinkles over the different test parameters and reference without intervention of the finger grippers (Ref., Experiment no. #6 from [8])

#### 4. Summary and Outlook

The systematic development of two concepts for the reduction of longitudinal wrinkles in calendered battery electrodes was presented. The validation tests have shown that the continuous roller concept with inclined conical and cylindrical spirally profiled rollers prevents longitudinal wrinkling under the conditions shown. The uncoated substrate of the guided electrode hits the deflection roller at a flatter angle. The required compensation in the form of longitudinal wrinkles is therefore reduced. The friction between the rollers and the uncalendered substrate induces an outward stretching force that is sufficient for the flatter uncoated substrate to prevent longitudinal wrinkling. However, the surface quality of the rollers must be improved in order to eliminate the imprints in the substrate. The finger gripper concept with the rubber contact surface was only able to change the shape of the longitudinal wrinkles. The height of the longitudinal wrinkles was reduced, while the longitudinal wrinkles formed directly at

the coating edge. Since manual stretching of the electrode showed promising results, it is assumed that the applied tensile force in the transverse direction is too low. Adapting the springdamper system, increasing the gripping surface and using a different contact surface could provide an improvement. Both systems are still very limited in their flexibility and automation and need further optimization.

In summary, longitudinal wrinkling in calendered electrodes can be reduced or even prevented by additional stretching units. The optimization of both concepts can help to reduce scrap in battery production in the long term. Due to the materialindependent approach, new and more sustainable battery systems can also be produced with reduced scrap in the future.

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#### Data Statement

The data associated with this article is available at: 10.5281/zenodo.10912128.

#### **CRediT Author Statement**

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