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Measuring Instruments for Characterization of Intermediate Products in Electrode Manufacturing of Lithium-Ion Batteries

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Electrode manufacturing is considered the core of lithium-ion battery cell production, with irreversible impacts on the electrochemical performance of the battery cell. The process chain is extensively complex, with a high number of interrelated parameters. The characterization of intermediate products in electrode manufacturing and the analysis of the correlations between process parameters and product properties can be considered a rigorous starting point to deepen process understanding and accelerate process optimization. Based on a holistic evaluation approach and a market analysis, this article provides a comprehensive overview of possible measuring instruments for intermediate products in electrode manufacturing, including the investment costs and inline/offline measurement strategy. The results can be used as a guideline for possible measuring instruments in electrode manufacturing, providing the foundation for in-depth process analysis. The findings demonstrate the current possibilities and highlight the need for further technological advancement in the field of metrology and digitalization.

1. Introduction

With the automotive industry striving to decarbonize the transport sector, the battery cell as a critical component in the value chain has been receiving exponentially greater attention over the last few years. For a breakthrough of the lithium-ion battery (LIB) technology in the automotive industry, certain quality and cost targets still need to be reached.[1,2] Parallel to the substantial material developments that aim to comply with quality requirements from the product perspective, advances in the production processes toward an efficient process chain are indispensable. Electrode manufacturing is considered the core of battery production, having irrevocable impacts on the battery cell's electrochemical performance.[2,3] With numerous interrelated parameters, the optimization of such a complex process chain requires an in-depth understanding of the process steps and their interactions that goes beyond a conventional trial-and-error approach. Over recent years, various studies have demonstrated the potential of data-driven methods to perform an in-depth and holistic analysis of interdependencies along the process chain.[4–9] A comprehensive understanding of the interdependencies between process parameters, intermediate products, and the final cell properties, achieved through data-driven analyses, can yield valuable insights into the entire production chain.[10] This serves as the foundation for implementing smart production, enabling efficient and well-informed decision-making at every stage of the production process chain.[11] Digitalization is the prerequisite for exploiting the full potential of such approaches. Ayerbe et al. reviewed the current status of digitalization in battery production and highlighted the opportunities and challenges that need to be addressed.[12] The main research domains are divided into four categories: 1) investigation of inline measurement technologies for data acquisition, 2) standardization of communication protocols for vendor-independent data access, 3) development of ontologies for the machine-readable abstraction of the domain knowledge, and 4) deployment of multiscale frameworks for handling and processing heterogeneous data streams.[12] Turetskyy et al. have focused on the fourth domain, presenting a framework for data acquisition and management.[13] This article aims to address the first research agenda and offers a comprehensive overview of measuring instruments in electrode manufacturing. The terms measuring system and measuring instrument are often used interchangeably. A measuring system is defined as a system that provides information about the physical value of a variable being measured.[14] The measuring system is seen as a collection of resources and components used to collect, transmit, process, and display the measuring data.[15] The term measuring instrument is commonly used to describe a measuring system adopted to conduct a measurement, whether it contains only one or many elements.[14,16] Following the definition provided by DIN 1319, these terms are adopted in this article.[16]

Reynolds et al. reviewed the possible measuring systems for data acquisition in the electrode coating process.[17] The measuring systems were evaluated based on their advantages and

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disadvantages for specific parameters such as coating thickness, mass loading, and coating defects.[17] Similarly, in a comprehensive review, Zhang et al. presented an overview of different drying mechanisms, including the key parameters, their impact on the electrode properties, and the possible in situ and ex situ measurement techniques.[18] Zanotto et al. highlighted the necessity of a digital twin to deal with the complexity of battery production.[19] The article contains an overview of the existing computation models in the LIB process chain and discusses the available acquisition techniques for experimental data.[19] In the extensive review, Zanotto et al. provided data specification for a set of parameters along the battery process chain, outlining their importance for the development and validation of models as well as their measurability and accuracy.[19]

A report from National Physical Laboratory (NPL) reviewed the measurement needs within the battery industry.[20] The report provides an overview of the primary measurement challenges and their priority along the battery life cycle, from materials through manufacturing to diagnostics and lifetime prediction. Major challenges in manufacturing include identifying the critical parameters influencing cell performance and enabling inline, fast, and noninvasive measurements of electrode and component properties during production.[20]

A tailored digitalization concept was introduced in a previous publication to prioritize parameters based on their relevance from the quality management perspective.[21] The result included a literature-based list of parameters, their relevance, and the complexity and effort involved in their digitalization. Following the previous work, this article evaluates possible measuring instruments that can be adopted to characterize intermediate products in electrode manufacturing. An overview of the process steps and the considered intermediate products is shown in **Figure 1**. Based on a holistic approach, this article aims to address the aforementioned challenges in the field of digitalization, explore the conventional inline measurement solutions, and provide an overview of possible measuring instruments that can be used as a guideline for process analysis and optimization. The details of the measuring principles of various solutions are beyond the scope of this article; comprehensive descriptions in this regard can be found in a number of handbooks and educational texts such as refs. [22–24].

2. Adopted Approach for Evaluation of Measuring Instruments

For the evaluation of measuring instruments, a three-step approach, according to Caulfield et al.,[25] is adopted. **Figure 2** provides an overview of the evaluation approach, divided into 1) definition of evaluation criteria, 2) search for measuring instruments, and 3) evaluation of measuring instruments. In the following, a brief description of each step is presented.

2.1. Definition of Evaluation Criteria

The definition of requirements and evaluation criteria makes a more systematic and efficient evaluation of measuring instruments possible.[25] The requirements should reflect the context of the studied use case. The relevant product parameters for the characterization of intermediate products are adopted from Haghi et al.[21] The production system's boundary conditions are decisive in selecting and evaluating measuring instruments. In this article, the boundary conditions are aligned with those defined in the previous work[21] and are briefly outlined in the following. The manufacturing readiness level (MRL) can be used as an indicator to describe the maturity of a production system and its processes.[26] The proposed solution in this article predominantly focuses on the pilot scale of LIB production with an MRL between 5 and 6, demonstrating the capability to produce prototype components in a production-relevant environment.[26] Given the critical role of the pilot scale as a bridge between the lab scale and industrial mass production,[27] it is essential to investigate the possible inline and offline measuring instruments for the LIB pilot lines. The findings can be used to optimize the quality control measurements from sampling to 100% inspection. For further details on the boundary conditions, refer to Haghi et al.[21]

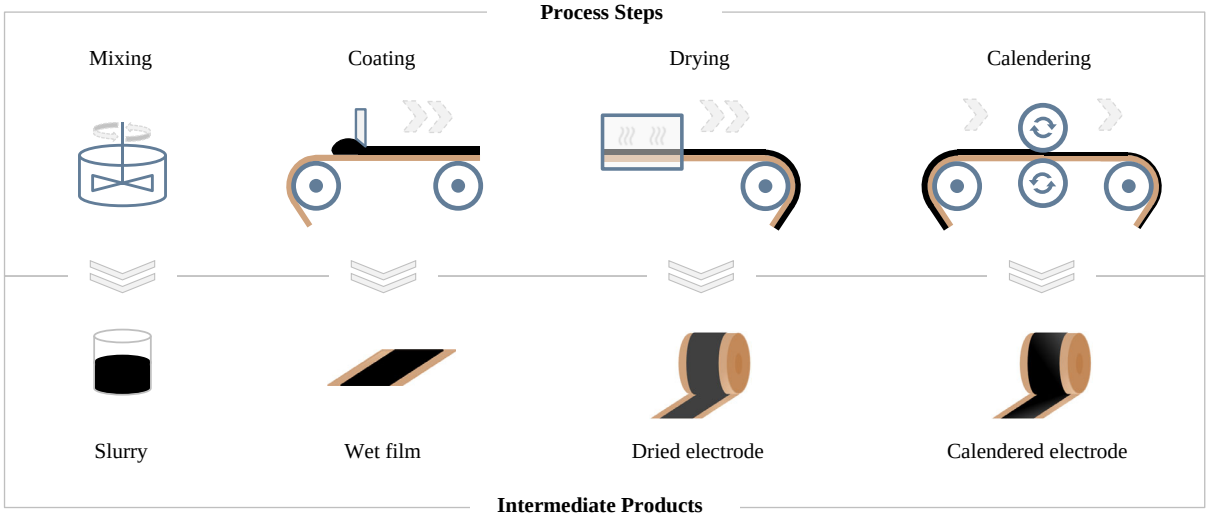

Figure 1. Overview of process steps and intermediate products in electrode manufacturing.

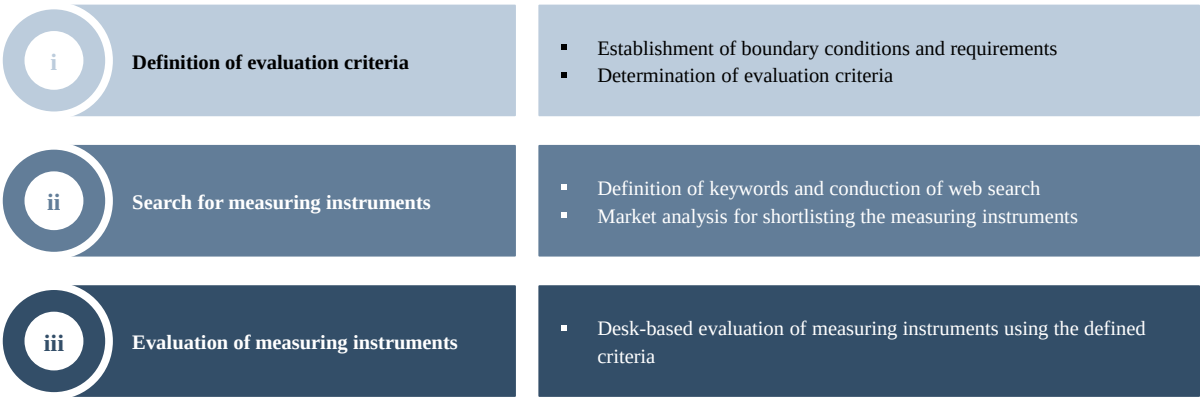

Figure 2. Adopted approach for evaluation of measuring instruments, according to Caulfield et al.[21]

The defined boundary conditions, such as relevant coating speed, are used to derive additional requirements for the measuring instruments. For example, for a resolution of 5 cm in the direction of the web at a coating speed of 15 m min^{-1}, an inline sensor requires a minimum sampling rate of 5 Hz. These requirements are taken into account during the market analysis in the next step.

Various standards have been published as guidelines to ensure the suitability of measuring systems for their intended use.[28–30] Squara et al. presented a comprehensive review of the key quality criteria in metrology provided by the International Bureau of Weights and Measures.[31] **Table 1** summarizes the selected evaluation criteria and their definitions for this study. It should be noted that the accuracy of a measurement is closely correlated with the measurement range. These criteria are adopted here as indicators for the possible measuring instruments. Concerning the measurement strategy, a distinction can be made between inline, at-line, and offline measuring systems.[32] An inline measuring system allows quality control of the product properties directly during the process, while at-line methods are based on the manual collection of samples with a short sample preparation.[32] The offline measuring systems are usually adopted in an analytical laboratory remote from the production line. They are typically associated with lengthy sample preparation and analysis time, making active process adjustments during production infeasible.[32,33] An example of an at-line method is a micrometer for measurement of electrode thickness via sampling at the production line, while a material testing machine can be considered as an offline method for measurement of electrode adhesion. In this article, at-line and offline measuring instruments are summarized under the offline category. The external conditions under which a measuring system is calibrated and used, such as temperature or vibration, can significantly impact the measurement procedure or results and should be taken into account.[28,34] It should be noted that there are some additional quality criteria, such as repeatability or reproducibility. However, the assessment of such factors demands extensive experimental analysis commonly seen as part of a measuring system analysis (MSA),[35] which falls out of this article's focus.

Table 1. Adopted evaluation criteria and their definitions.

Evaluation criteria	Definition
Measurement range	Minimum and maximum values which can be measured
Accuracy	Closeness of the agreement between the outcome of a measurement and the true value of the measurand[31]
Capital expense	Costs associated with the acquisition of the measuring instrument
Measurement strategy	Differentiation between inline with real-time data collection during the process and at-line/offline measurement based on manual discontinuous sampling[32]

2.2. Search for Measuring Instruments

In the second step, a web search was conducted to identify available measuring instruments for battery electrode manufacturing. In case commercialized measuring instruments were already known from the experimental sections of various publications, these were integrated as keywords into the web search (see **Table 2**).

Lopez-Vega et al. proposed four search paths to identify innovative technologies based on the two dimensions of search space and search heuristics.[36] In this article, the search space is evaluated as local, limited to measuring instruments compatible with battery manufacturing. The search is conducted based on a cognitive rather than experimental approach. Therefore, the analysis undertaken in this study can be classified as the "sophisticated path" category defined by Lopez-Vega et al.[36] This category aims to look for short-to-medium-term insights into possible technological solutions.[36] Hence, by shortlisting the possible measuring instruments, only those with a technology readiness level (TRL) higher than five are considered, excluding the prototype solutions presented in academia.[37]

Table 2. Overview of the categories used for the web search with exemplary keywords.

Category	Exemplary keywords
Specification of the use case	Electrode manufacturing
	Lithium-ion battery production
Product property	Wet film thickness
Possible measuring instruments	Laser triangulation
	Confocal sensor

The relevant specifications of electrode manufacturing, such as associated ranges, have also been taken into account to ensure the compatibility of the measuring system with battery production. The market analysis was conducted in the time interval between September and November 2022, resulting in the identification of more than 40 national and international measuring instrument suppliers. For the evaluation of measuring instruments, the suppliers were contacted in the next step.

2.3. Evaluation of Measuring Instruments

The last step involved a desk-based evaluation of the measuring instruments. For this purpose, the identified suppliers were contacted. Product catalogs and datasheets were used to gather the technical information required to evaluate the measuring instruments. In terms of investment costs, the data were first collected for all the different measuring instruments. Subsequently, based on the existing ranges, superclasses were defined, dividing the investment costs into a total of six categories; these are summarized in **Table 3**.

3. Results of the Evaluation Approach

Following a brief description of the evaluation approach in Section 2, this section outlines the results of the adopted approach. In addition to the evaluation criteria defined in the first step, the importance of the product parameter from the quality management perspective is integrated based on the results from Haghi et al.[21] This can be used as an additional indicator for decision-making concerning digitalization and measurement solutions.

3.1. Measuring Instruments for Characterization of Slurry

As the first step in the battery production chain, the mixing process has an immense impact on downstream processes, the electrode's microstructure properties, and the battery cell's electrochemical performance.[38] From the digitalization perspective, one of the main challenges is the lack of inline measuring systems during the mixing process with conventional discontinuous mixing technologies.[21] The rheological properties of the slurry, as the final product of the mixing process, are critical for the subsequent coating step. A rotational rheometer can be used to measure viscosity at lower shear rates, while a capillary rheometer is ideally suited to determine the dynamic viscosity at higher industry-relevant shear rates. Besides viscosity, viscoelastic properties of the slurry—such as storage and loss modulus—can be determined by the rheometer using amplitude or frequency sweep in an oscillatory test.[39] It should be noted that the sample's temperature can influence the measurement result. Hence, it is essential to include an equilibrium period of, for example, 5 min before conducting the measurement to ensure the correct temperature of the sample.

Table 3. Overview of the defined cost categories and the associated aggregated range.

Capital expense	Aggregated range
Category 1	Less than €2000
Category 2	€2000–€20 000
Category 3	€20 000–€100 000
Category 4	€100 000–€200 000
Category 5	€200 000–€500 000
Category 6	More than €500 000

Another critical characteristic is the slurry's homogeneity, which is defined by the uniform distribution of the components throughout the batch, especially the conductive agent. The particle size distribution and the tendency of the particles toward agglomeration and sedimentation can be used as indicators of the slurry's homogeneity and stability. Two measurement principles are deployed for the particle size and particle shape analyzer: dynamic light scattering and laser diffraction. While the former can be used to measure nanoparticles, the latter covers a more extensive particle size range. As a common conductive agent, carbon black has a particle diameter of 20–60 nm.[40] The small particle size imposes a measurement challenge. To be able to conduct a precise, reliable particle size analysis, mapping the nanoparticles instead of their agglomerates, Dreger et al. proposed an efficient sample preparation method.[41] As an alternative, a zeta potential meter can be adopted to measure the size and surface charge of the particles.[42] A higher zeta potential can indicate a more stable slurry that is less prone to agglomerate formation.[43] The fineness of grind gauge, also known as the Hegman gauge, is a simple instrument that can be used as a quick check, indicating the fineness of the slurry and the presence of agglomerates.[44] The sedimentation rate can be estimated based on the rheology measurement and the Stroke law.[45]

The density of the slurry can be precisely measured by a digital density meter based on an oscillating U-tube principle. The solid content of the slurry can be calculated through the density of the components and the final slurry. As an alternative, a moisture analyzer can be used to determine the solid content.

Surface tension is another important slurry property that should be monitored, as it can impact cracking issues, especially in the case of thick electrodes.[46] The surface tension is closely correlated with the contact angle, representing the shape that a liquid takes on a solid surface.[47] The surface tension can be measured by a force tensiometer or indirectly calculated with an optical tensiometer by analyzing the pendant drop shape. Two methods are adopted by the force tensiometer: the Du Noüy ring and the Wilhelmy plate method.[48] It should be noted that the surface tension is not independent of the slurry's temperature.[49] Therefore, the temperature should be considered in the sample preparation and interpretation of the measurement result. For this purpose, a temperature control unit is usually integrated into the tensiometers available in the market.

The pH value of the slurry is another crucial parameter to consider, especially as further progress is made in aqueous processed cathodes.[50,51] This parameter can be measured using a benchtop pH meter.

During the market analysis, a slurry analytical system capable of measuring the slurry's electrical conductivity was identified. The measuring system is expected to be available on the European market by the end of 2023. As an alternative and a

more common solution, the resistivity of the electrode can be measured by a resistance measurement system (see Section 3.3). **Table 4** provides an overview of the results of the desk-based market analysis. The measurement range and accuracy are included, as reported by the suppliers. In combination with capital expenditure, these specifications can be used as indicators for possible measuring instruments.

3.2. Measuring Instruments for Characterization of Wet Film

The coated wet film can serve as a gauge for the quality of the dried electrode. The wet film thickness and mass loading are highly correlated to the final electrode mass loading. Different technologies are adopted to measure these properties inline. Possible inline measuring systems for the wet film thickness include laser triangulation,[52,53] confocal chromatic sensor,[54] or white light interferometer.[18,55–57] It should be noted that the measurement range and accuracy reported for these systems are affected by their distance from the object and should be interpreted as approximate reference values. Among the measuring instruments, laser triangulation can cover a broader measurement range.[58] An overview of typical accuracy at different operating distances of the thickness measuring systems—which consequently impact the measurement ranges—can be found in ref. [58]. Laser triangulation systems can scan over a wide area, hence they can be used to measure the thickness across the coating width.[17,59] Due to this capability, the laser triangulation system can also be used for inline monitoring of the coating width during the process. If the coating width exceeds 25 cm, two measuring systems should be included for this purpose. In the case of other measuring instruments, the thickness sensor can be mounted on a traversing system, allowing a thickness profile obtained across the coating width. It should be noted that the traversing setup also impacts the measuring instrument's sampling rate. The influence of the electrode web's vibration on the measurement results is another crucial aspect to be considered when integrating a thickness measuring system. Therefore, such systems are best mounted in combination with a roller in the coater. In the case of the laser triangulation and confocal sensors, another possibility to increase the measurement accuracy is to incorporate a C-Frame or O-Frame setup—enabling measurements from both sides of the electrode. Mohanty et al. have analyzed such a setup using two laser triangulation systems and quantified the expected errors that might occur due to various factors such as misalignment and vibration.[60] The reflective surface of the wet film might also cause a certain degree of fluctuation in the measurement results.

The aforementioned challenges, such as vibration and surface reflection, can be avoided by measuring the wet mass loading instead of the film thickness. Four technologies are deployed for inline characterization of the electrode mass loading: ultrasound, near-infrared (NIR), beta, and X-ray transmission. It should be noted that these inline solutions have a different measurement principle compared to common offline measuring instruments such as strain gauge or force restoration scales. Hence, a calibration step should usually be considered while configuring the inline measuring instruments. For this purpose, samples can be taken from the electrode and characterized offline, for example, using an analytical scale and micrometer. The mass loading determined offline can then be compared with the value measured by the inline measuring system. Based on this, the final target values for the inline measuring instrument are determined. Additionally, it should be noted that these inline systems rely on comparing two materials—electrode coating and substrate foil—with different densities to determine the mass loading.[17] Hence, the measurement results may be subject to a certain degree of error.[17] To accurately map the electrode's mass loading and avoid large tolerances, it is suggested to integrate three measurement units into the coater, measuring the substrate foil, the wet film, and the dried electrode individually. A comprehensive comparison of different measurement technologies for mass loading systems is presented by Reynolds et al.[17] The possible limitations include the restricted traversing speed of radiation-based methods, which may result in constraints on the industrial scale and the need for additional safety measures.[17]

The coating edge elevation, as a quality-critical undesirable phenomenon in the slot-die coating, can be characterized using a laser triangulation system.[52,61] Two dimensions are relevant for the characterization of this phenomenon: the elevation height and its expansion across the coating width.[61] The measurement solutions for mass loading can also be used for this purpose.

Table 4. Overview of intermediate product parameters in the mixing process and the possible measuring instruments.

Parameter	Importance[a)]	Measuring instrument	Measurement range	Accuracy	Capital expense	Measurement strategy
Rheological properties	■	Rheometer	1–10^9 mPas[b)]	1–9%	Category 4	Offline
Homogeneity (particle size, agglomerate)	■	Particle size and shape analyzer	0.3–1 mm	<2%	Category 3	Offline
		Zeta potential analyzer	±500 mV	–	Category 3	Offline
		Hegman gauge	0–100 µm	5%	Category 1	Offline
pH value	■	pH meter	−2 to 20	0.002	Category 1	Offline
Surface tension	■	Tensiometer	1–2000 mN m^{-1}	–	Category 3	Offline
Slurry density	■	Density meter	−3 mg cm^{-3}	0.005 mg cm^{-3}	Category 2	Offline
Solid content	■	Moisture analyzer	<100 g[c)]	0.01%	Category 1	Offline

[a)]Importance: ■ highly important parameter; ■ less important parameter. [b)]The possible range for dynamic viscosity. [c)]Amount of sample.

Another quality-critical aspect that can have an irreversible impact on cell performance is the electrode's defects, such as pinholes or stripes.[62,63] To ensure that all potential defects arising during the coating and drying process are detected, the measuring systems are most often installed after the drying section of the coating line. Hence, these are listed in Section 3.3. **Table 5** summarizes the possible measuring instruments in the electrode coating process.

3.3. Measuring Instruments for Characterization of Dried Electrode

Similar to the coating process, the electrode thickness and mass loading are considered the main product parameters in the drying process. The same measuring systems listed in Section 3.2 can be adopted for the dried electrode.

The drying condition, determined by the web speed and the temperature profile, in conjunction with the electrode thickness, can impact the electrode's microstructure[64–66] and its mechanical properties.[67] Indentation tests, on micro- and nanoscales, can be used to characterize the mechanical properties of the electrode.[68] Ideally, a noise-free environment is recommended for the nanoindentation test, with a certain degree of vibration control. Additionally, pull-off[69,70] and 90° peel tests[71] can be performed on a material testing machine to evaluate the electrode's adhesion as a crucial mechanical property. Sample preparation is an important aspect to be considered in the pull-off test to avoid possible inadvertent failure mechanisms.[67] In the pull-off test, a round specimen is preferred to a rectangular one due to the uniform areal force distribution, which leads to more accurate results. Haselrieder et al. have established a standardized pull-off test, including the parameter setup to measure the adhesion strength.[67]

The material testing machine can also be used to measure the flexural strength of the electrode using a two-point bending test. As a qualitative alternative to characterize the electrode's bending flexibility, a cylindrical mandrel bend tester can be used for a quick check. From the production perspective, this parameter is important in the subsequent roll-to-roll process steps, while from the product point of view, it can be used as a quality indicator to characterize the suitability of the electrode for cylindrical cells. The bend tester is available in a variety of cylindrical sizes. A standard core diameter of a coil in battery production is between 80 and 152 mm. The electrodes' bending flexibility and crack formation are determined by wrapping the electrode around the mandrel. A set of indicators, such as the shape and distribution of the cracks formed, can be used as evaluation criteria.[72]

The drying process is one of the most complex process steps in battery production, which can impact the homogeneity of the electrode on the microstructure scale and, consequently, the final cell performance.[73] Extensive research has been dedicated to analyzing this process step and the strongly associated undesirable effect of binder migration.[74–79] Different offline measuring systems can be used to characterize the morphology of the electrode and approximately map the binder distribution. With the help of a scanning electron microscope (SEM) and energy-dispersive X-Ray analysis (EDX), the distribution of components within the electrode can be investigated.[80] Zheng et al. presented an extensive list of different offline techniques for the characterization of the drying process, such as Raman spectroscopy and atomic force microscopy.[18] It should be noted that these techniques are still considered indirect measurement solutions, and some may fail to meet the requirements due to factors such as specific formulation with a certain amount of binder concentration.[81] Another possibility is the application of eddy current as an inline nondestructive measuring system using electromagnetic induction. Such systems can commonly be used to measure the coating's resistance and electrical anisotropy.[82,83] However, the applicability and transferability of this characterization solution have not yet been fully demonstrated in battery production.

In addition to edge elevation, compositional inhomogeneity, and microstructural heterogeneity, other defects, such as pinholes or agglomerates, can occur during the coating and drying processes. A comprehensive review of different defect types with their influence on the electrochemical performance of the battery cell is provided by Du Baret de Limé et al.[62] Infrared (IR) thermography or visual inspection systems using a camera can be adopted to identify electrode surface defects.[63] Depending on the pixel size and optics, such systems can have resolutions in the µm range. The available visual inspection systems in the market also provide data-driven image processing and classification for automated defect detection. It should be pointed out that the

Table 5. Overview of intermediate product parameters in the coating process and the possible measuring instruments.

Parameter	Importance[a)]	Measuring instrument	Measurement range	Accuracy	Capital expense	Measurement strategy
Wet film thickness	■	Laser triangulation[b,c)]	< 10 mm[d)]	$\pm$ 2 µm	Category 3	Inline
		Confocal chromatic sensor	< 1.5 mm[d)]	–	Category 3	Inline
		White light interferometer	< 4 mm[d)]	–	Category 3	Inline
Wet film mass loading[b,e)]	■	Ultrasound system	1–900 g m^{-2}	0.5%	Category 5	Inline
		NIR system	1–300 g m^{-2}	0.1%	Category 4	Inline
		Beta gauge	2.5–900 g m^{-2}	–	Category 3	Inline
		X-Ray system	5–900 g m^{-2}	0.25%	Category 4	Inline

[a)]Importance: ■ highly important parameter; ■ less important parameter [b)]The measuring instrument can also be used to monitor the coating edge profile. [c)]The measuring instrument can also be used to monitor the coating width (accuracy for the coating width: $\pm$ 20 µm). [d)]The reference value for a working distance of approx. 2–3 cm [e)]The costs are based on three measurement units: measuring the substrate foil, the wet film, and the dried electrode.

cost of such systems closely depends on the specifications of the production line, such as the coating width and the web speed. Similar to the thickness measuring instruments, the vibration aspect should also be considered by the integration of the camera systems. Hence, these systems are preferably mounted in combination with a roller or between two closely positioned rollers in the coater.

Another parameter that can be used to characterize the electrode after the drying process is electronic resistance.[69] It should be noted that electronic resistance can also be influenced by the mixing and calendering processes.[84] The resistivity of the electrode can be measured by a resistance measurement system using a two- or four-point probe testing method. The latter is preferred due to higher accuracy. Given the geometric properties of the sample and the current collector resistivity, the measuring instrument can distinguish between composite layer resistance and interface resistance between the composite electrode and the current collector.

Mercury intrusion porosimetry (MIP), as a powerful destructive method, is used to evaluate the porosity and pore size distribution within the electrode composite.[85] Sample preparation is an essential aspect of the MIP analysis. Radloff et al. examined the effects of the sample's geometry and mass on the MIP result.[86] As the calendering process ultimately determines porosity, this measuring system is conventionally applied to the calendered electrode. However, it can also be adopted to characterize uncalendered electrodes to thoroughly investigate the effects of drying conditions or slurry formulation, which have been shown to impact porosity.[87,88] A gas pyrometer can also be adopted to measure the skeletal density of the electrode components and calculate porosity.[89,90] Alternatively, the average crystallographic density of electrode components is used to estimate porosity.[91] Based on the assumption of homogeneous density, the latter approach can be used for indirect inline measurement of porosity using thickness and mass loading measuring systems.

Tortuosity is another essential metric that is considered to impact the electrolyte's diffusivity inside the electrode's pores and thus cell performance.[92] Various experimental methods have been dedicated to measuring tortuosity, from tomography to electrochemical setup using impedance spectroscopy.[93–97] Tjaden et al. provided a comprehensive overview of tortuosity estimation approaches.[98] The variety of the experimental methods, combined with the wide range reported for tortuosity in the literature, implies that the experimental techniques related to tortuosity estimation may not have yet been honed[39] and are strongly influenced by experimental artifacts.[94]

In recent years, efforts have been made to find alternative solutions for cost-effective and environmentally sustainable battery production. One viable approach is enabling the aqueous processing of cathode material and the elimination of *N*-methylpyrrolidone (NMP) in the process chain.[99] Given this endeavor and the existing solvent for anode formulations, moisture content can be regarded as a relevant product parameter that can be used as an indicator to evaluate and adjust the drying process parameters. Karl Fischer titration is a conventional offline lab-scale method to determine the water content in a sample using volumetric or coulometric titration.[100] While the former is best suited for the determination of water content in higher ranges up to 100%, the latter is suitable for ranges below 5%. Near-infrared hyperspectral imaging (NIR-HSI) system is an inline nondestructive alternative to characterize moisture content.[101] While such systems have received increasing attention in other sectors, such as the food industry,[102–104] they are still not well established in battery production. **Table 6** outlines the possible measuring instruments for the characterization of the dried electrode.

3.4. Measuring Instruments for Characterization of Calendered Electrode

The majority of the parameters described in Section 3.3 for the drying process are also considered in the calendering process. However, as their relevance from the quality management perspective within each process might vary, they are shortly described and listed separately in this section. Electrode thickness is used as an indicator to achieve the desired compression rate in the calendering process.[105] The same measuring instruments outlined in Section 3.3 for the thickness of uncalendered electrodes can be used in the calendering process.

The calendering process defines the electrode's final porosity, which directly impacts electrochemical cell performance.[106] As described in Section 3.3, offline methods such as MIP can be used to measure the porosity of the calendered electrode.[85] Electrode thickness and mass loading measurements can be adopted as an indirect inline alternative. As the mass loading of the electrode is not affected during the calendering process, it is possible to consider sensor fusion using the mass loading data collected in the previous process step, and the electrode thickness measured inline after calendering. An accurate data mapping can be ensured through a tracking and tracing system.[107–110]

Electronic resistance is another critical product property that can be affected by the calendering process. The higher compaction realized through the calendering process leads to increased energy density and improvement in the electronic conductivity of the electrode. The offline resistance measurement system can be used to characterize the calendered electrode.

In contrast to electronic resistance, tortuosity usually increases at higher compaction rates as ionic pathways tend to get longer. Tortuosity, therefore, defines the ionic resistance within the electrode and can be indirectly measured by creating a high-resolution 3D model of the electrode.[18] The FIB-SEM and X-Ray nano-CT are possible techniques for developing such 3D models.[98] A potentiostat can be used to conduct electrochemical impedance spectroscopy and characterize the electrode's ionic conductivity[106] and tortuosity.[94,96]

The mechanical properties of the electrode are also affected by the calendering process. Excessive compression leads to particle breakage, resulting in increased moisture adsorption and reduced rate capability.[111] A well-parameterized calendering process positively impacts the adhesion, cohesion strength, and the electrode's homogeneity.[70,112] Other important parameters are the elastic and plastic deformability within the electrode. The deformability can be destructively evaluated using micro- or nanoindention measurements.

Table 6. Overview of intermediate product parameters in the drying process and the possible measuring instruments.

Parameter	Importance[a)]	Measuring instrument	Measurement range	Accuracy	Capital expense	Measurement strategy
Electrode thickness	■	Laser triangulation	<10 mm[b)]	± 2 µm	Category 3	Inline
		Confocal chromatic sensor	<1.5 mm[b)]	–	Category 3	Inline
		White light interferometer	<4 mm[b)]	–	Category 3	Inline
Electrode mass loading[c)]	■	Ultrasound system	1–900 g m^{-2}	0.5%	Category 5	Inline
		NIR system	1–300 g m^{-2}	0.1%	Category 4	Inline
		Beta gauge	2.5–900 g m^{-2}	–	Category 3	Inline
		X-Ray system	5–900 g m^{-2}	0.25%	Category 4	Inline
Electronic resistance	■	Resistance measurement system	<12 × 10^6 Ω	0.5%–3%	Category 3	Offline
Mechanical properties (e.g., adhesion, bending flexibility)	■	Material testing machine	0.5–5 kN	–	Category 3	Offline
		Microindenter	Force: max. 30 N Depth: max. 1 mm	–	Category 5	Offline
		Nanoindenter	Force: 0.1 mN–1 N Depth: 200–600 µm	–	Category 5	Offline
		Cylindrical mandrel bend tester	–	–	Category 1	Offline
Morphology and homogeneity	■	Scanning electron microscope (SEM)	10 nm–200 µm	–	Category 6	Offline
		Atomic force microscope	10–200 µm	–	Category 4	Offline
		Optical microscope	>2 µm	–	Category 3	Offline
		Raman spectrometer	0.5–1300 µm	–	Category 5	Offline
Porosity	■	Mercury intrusion porosimetry[d)]	3.5 nm–1100 µm	–	Category 3	Offline
		Gas pycnometer[d)]	0.7 nm–100 µm	–	Category 3	Offline
		Combination of coating thickness and mass loading	–	–	Depending on individual instruments	Inline
Tortuosity[e)]	■	Focused ion beam microscope (FIB-SEM)	10 nm–1 cm	–	Category 6	Offline
		X-Ray nanocomputed tomography (CT)	1 nm–1 cm	–	Category 6	Offline
		Potentiostat[f)]	–	–	Category 3	Offline
Quality of electrode surface	■	IR thermography	–	–	Category 4	Inline
		Machine vision for optical inspection	–	–	Category 4	Inline
Moisture content (H_2O)	■	Karl Fischer titrator	1–50 000 ppm	–	Category 3	Offline
		NIR-Hyperspectral imaging system	0.1%–40%	–	Category 3	Inline

[a)]Importance: ■ highly important parameter; ■ less important parameter [b)]The reference value for a working distance of approx. 2–3 cm [c)]The costs are based on three measurement units: measuring the substrate foil, the wet film, and the dried electrode. [d)]The range reported for the pore size. [e)]Only indirect measurement is possible. [f)]The cost is based on an 8-channel potentiostat/galvanostat.

Günther et al. have classified the possible defects in the calendering process into three categories: geometrical, structural, and mechanical.[113] Similar to the drying process, high-precision imaging methods can be adopted to characterize the electrode given the structural defects and inhomogeneity based on samples. In view of the geometrical defects, such as corrugations at the coating edge or the camber effect, efforts have been made to detect such defects inline using laser triangulation systems.[114,115] **Table 7** summarizes the intermediate product parameters and the possible measuring instruments in the calendering process.

The final results of the systematic evaluation approach are visualized in **Figure 3**. For the distribution between inline and offline measurement strategies, offline measuring instruments such as the material testing machine or SEM were considered only once since the same system can be deployed to characterize different intermediate products. The various technologies for inline measurement of mass loading in the coating and drying process have also been consolidated, as the investment costs were based on three systems measuring the substrate foil, the wet film, and the dried electrode.

The results show that there is still a set of intermediate product parameters that can be characterized only using offline measuring instruments. This distribution demonstrates the underlying need for further technology development regarding inline measuring solutions, particularly for the parameters deemed highly relevant from the quality management point of view.[21] The inline solutions require a capital investment higher than EUR 20 000 per measuring instrument. The highest investment categories include merely offline solutions such as SEM

Table 7. Overview of intermediate product parameters in the calendering process and the possible measuring instruments.

Parameter	Importance[a)]	Measuring instrument	Measurement range	Accuracy	Capital expense	Measurement strategy
Electrode thickness	■	Laser triangulation	< 10 mm[b)]	$\pm$ 2 µm	Category 2	Inline
		Confocal chromatic sensor	< 1.5 mm[b)]	–	Category 2	Inline
		White light interferometer	< 4 mm[b)]	–	Category 2	Inline
Mechanical properties (e.g., adhesion, elastic and plastic deformation)	■	Material testing machine	0.5–5 kN	–	Category 3	Offline
		Microindenter	Force: max. 30 N Depth: max. 1 mm	–	Category 5	Offline
		Nanoindenter	Force: 0.1 mN–1 N Depth: 200–600 µm	–	Category 5	Offline
		Cylindrical mandrel bend tester	–	–	Category 1	Offline
Morphology and homogeneity	■	Scanning electron microscope (SEM)	10 nm–200 µm	–	Category 6	Offline
		Atomic force microscope	10–200 µm	–	Category 4	Offline
		Optical microscope	>2 µm	–	Category 3	Offline
		Raman spectrometer	0.5–1300 µm	–	Category 5	Offline
Electronic resistance	■	Resistance measurement system	$<12 \times 10^6$ Ω	0.5–3.0%	Category 3	Offline
Porosity	■	Mercury intrusion porosimetry[2]	3.5 nm–1100 µm	–	Category 3	Offline
		Gas pycnometer[c)]	0.7 nm–100 µm	–	Category 3	Offline
		Combination of coating thickness and mass loading	–	–	Depending on individual instruments	Inline
Tortuosity[d)]	■	Focused ion beam microscope (FIB-SEM)	10 nm–1 cm	–	Category 6	Offline
		X-Ray nano-computed tomography (CT)	1 nm–1 cm	–	Category 6	Offline
		Potentiostat[e)]	–	–	Category 3	Offline
Ionic conductivity	■	Potentiostat[e)]	–	–	Category 3	Offline
Quality of electrode surface	■	IR thermography	–	–	Category 4	Inline
		Machine vision for optical inspection	–	–	Category 4	Inline
		Laser triangulation	<10 mm[b)]	$\pm$ 2 µm	Category 3	Inline
Moisture content (H_2O)	■	Karl Fischer titrator	1–50 000 ppm	–	Category 3	Offline
		NIR-Hyperspectral imaging system	0.1–40%	–	Category 3	Inline

[a)]Importance: ■ highly important parameter; ■ less important parameter. [b)]The reference value for a working distance of approx. 2–3 cm. [c)]The range reported for the pore size. [d)]Only indirect measurement is possible. [e)]The cost is based on an 8-channel potentiostat/galvanostat.

and Raman spectrometer. The drying process accounts for the majority of product parameters; however, there is an overlap between the drying and calendering processes in terms of product properties. Porosity, as an example, is ultimately defined during the calendering process. Nevertheless, the characterization of uncalendered electrodes concerning porosity might be of interest, depending on the objective of the analysis.

It is important to highlight that the results presented in this study primarily focus on investment costs. While offline solutions can be acquired and seamlessly integrated into the pilot line without causing interruptions in the ongoing production, incorporating inline solutions during the brownfield phase may lead to additional expenses. Certain inline measuring instruments, such as those listed for electrode thickness measurement, can be integrated with restricted downtime. However, incorporating more complex measuring instruments, such as mass loading systems, may necessitate additional time and resources.

4. Conclusion

This article has focused on evaluating the possible measuring instruments for the characterization of intermediate products in lithium-ion electrode manufacturing using a holistic and systematic approach. Based on a market analysis, possible measuring systems, including the measurement range, measurement strategy, and capital expense, were outlined to provide both researchers and practitioners with a guideline for the characterization of intermediate products. In combination with the tailored digitalization concept, which includes a prioritization of parameters from the quality management perspective,[21] the results can serve as a baseline for decision-making concerning digitization initiatives in electrode manufacturing.

The findings regarding possible inline and offline methods indicate the need for further development of inline measuring systems. Priority should be given to the parameters ranked high from the quality management perspective.[21] While plausible yet

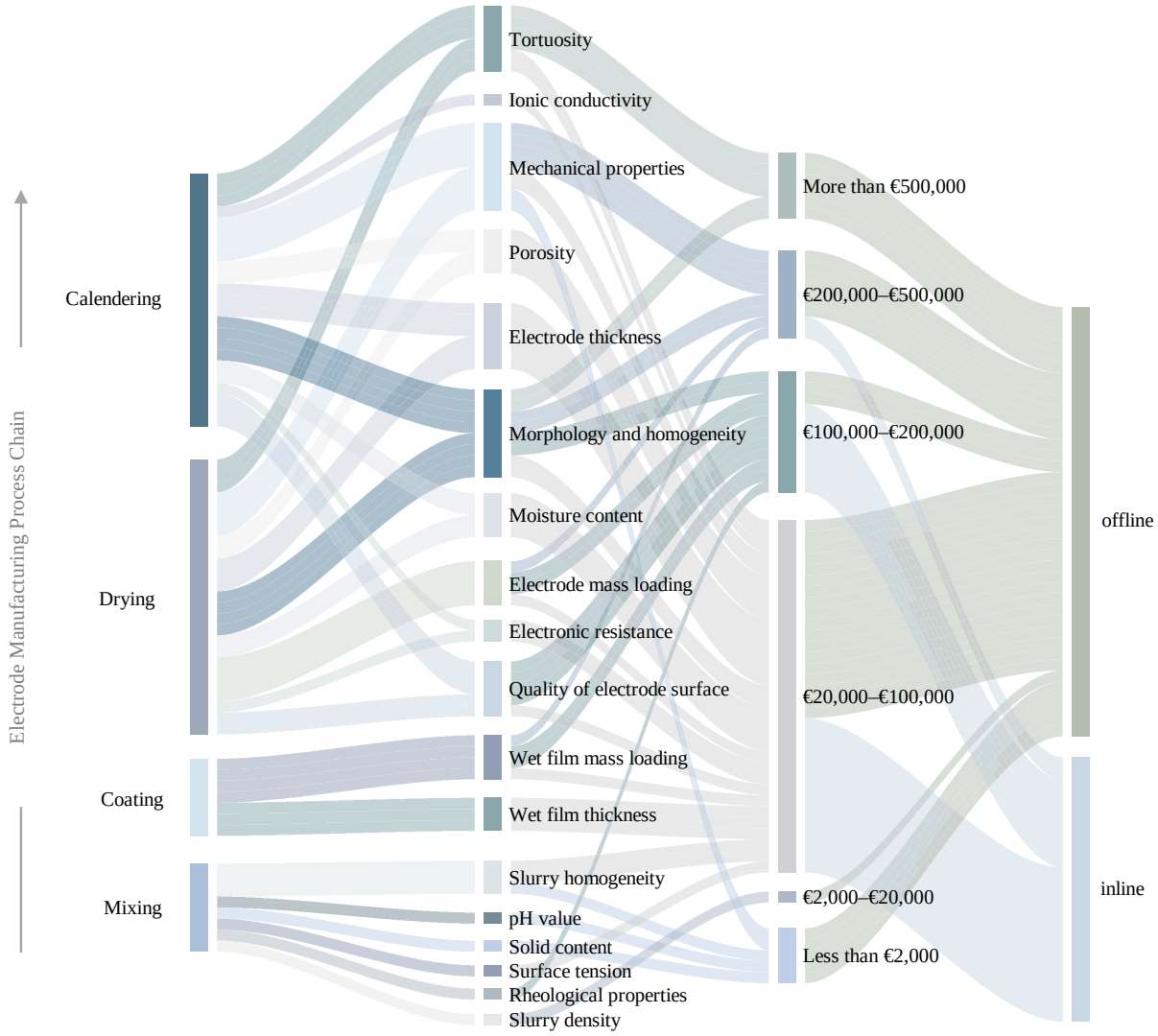

Figure 3. Sankey diagram illustrating from left to right: the process steps, intermediate product parameters, the associated costs of the measuring instruments for the product parameters and the measurement strategy. The widths of the links are proportional to the number of measuring instruments.

elaborate solutions exist, for example, to characterize the porosity of the electrode inline based on data fusion using the mass loading and thickness measuring systems, finding nondestructive inline measurement solutions for a set of parameters, such as adhesion, is nearly unattainable. To address such cases, two approaches are currently being pursued in the research community: indirect characterization of the electrode properties using novel inline solutions such as spectrophotometry[116] or application of data-driven models. Based on the results of the offline measuring instruments and the inline collected data, machine learning models can reveal the interdependencies between significant parameters, such as adhesion, mass loading, and drying temperature.[117] Ultimately, a data-driven smart production system is expected to impact the frequency and sampling size required for the application of offline measuring systems. The adoption of digital technologies in combination with data-driven approaches in battery pilot production lines can enhance process understanding, accelerate the scale-up of novel material generations, and pave the way toward quality-oriented, cost-efficient, sustainable battery cell production.

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

S.H.: Conceptualization, methodology, investigation, visualization, writing—original draft. M.L.: Investigation, writing—original draft. A.M.:

Investigation, writing—review and editing. R.D.: Funding acquisition, supervision, writing—review and editing.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

digitalization, electrode manufacturing, lithium-ion batteries, measuring systems, metrology, sensor technology

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