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Identification of Relevant Parameters for Traceability in the Continuous Mixing Process in Battery Cell Production

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The growing demand for lithium-ion batteries in electric vehicles pose challenges, particularly for the use of critical materials such as cobalt in electrode slurry mixing. Due to regulatory requirements such as the battery passport, an accurate traceability system is mandatory. However, there is no solution for the continuous mixing process, which is a major challenge in electrode production. This article describes an approach to solve this problem. Based on the results of a literature review, a design structure matrix is constructed to analyze parameter influences. The overall result is a list of relevant parameters that should be recorded in the continuous mixing process in order to characterize the mixing process.

1. Introduction

The shift toward zero-emission and sustainable mobility is driving demand for electric vehicles and lithium-ion batteries.[1] Car manufacturers are increasingly concentrating on the production of their own battery cells.[2] The key challenges include reducing costs, minimizing scrap, and at the same time improving quality.[2] For this reason, particular attention is paid to resource-saving production and process optimization in the production of battery cells. The idea of resource-efficient battery cell production is also reflected in the new regulation for battery production in the European Union.[3] The European Union supposes that battery cell production should be focused on optimizing performance, durability, and safety. Therefore, from the 1st of January 2027 a battery information system will be mandatory, which will allow to collect and use information and data on individual batteries placed on the market more efficiently. Binding quotas are also set for minimum quantities of materials recovered from used batteries; for example, for cobalt, copper, lead, and nickel, 90 percent by 2027 and 95 percent by 2031, and for lithium 50 percent by 2027 and 80 percent by 2031.[3] This requires targeted and specific traceability, which is based on selected parameters, along the supply chain, but also in the battery cell production.

It is already possible to record the information and data of intermediate products in battery cell production from the coating process step onward and assign them to the individual cells.[4–8] The mixing process in general and the continuous mixing process in particular pose a great challenge for traceability. This is due to the mixing of different materials to a slurry paste, the mixing of multiple batches of slurry in the buffer tank prior to the coating process, and the unknown interdependencies between the individual parameters.[9] It will only be possible to link information from the process, the product and the physical world perfectly once this challenge has been solved. In the mixing process, traceability is currently only possible for batch processes by attaching radio frequency identification (RFID) tags or tracking codes to the slurry containers.[4,5,7] However, inaccuracies and product inhomogeneities cannot be avoided due to the large quantities of slurry that are stored in a container. There is currently no way to ensure traceability in continuous slurry mixing - neither general nor parameter-specific.[5,7] In addition, it is not yet clear which parameters must be recorded in a traceability system for the continuous mixing process because these have the greatest influence or best characterize the process.

2. Background

In general, battery cell production is divided into the three main processes of electrode manufacturing, cell assembly, and cell finishing. The electrode manufacturing process is again divided into the process steps of mixing, coating, drying, and calendering.[2,10]

The first process step of the electrode manufacturing is the mixing process of the electrode slurry. Here, the active materials, binder and conductive agent, which commonly use carbon black (CB), are diluted in a solvent.[11] The aim of mixing is to break up existing material agglomerates and to provide a homogeneous slurry with specific viscosity.[2,12] The mixing process is divided into wet mixing and dry mixing. Wet mixing process literally means preparing the slurry by mixing and dispersing the materials in the presence of solvent solution, while dry mixing is in

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the absence of liquids. Usually, active material and conductive agent are mixed using dry mixing technique, following with wet mixing technique by adding solvent and binder.[13] Dry and wet dispersion processes determine the size, morphology, distribution, and structural arrangement of the conductivity additives. By mixing conductive agent with binder, it will promote a gel-like slurry. Meanwhile, mixing active material and conductive agent will increase the surface area and slurry viscosity. Mixing techniques, instrumentation, intensity, duration, and sequences are among the most important factors in determining slurry quality.[12] The mixing process is of particular importance for several reasons: Firstly, the mixing largely and irrevocably determines the electrochemical performance of the battery cell and its properties.[14] Furthermore, the mixing process affects the downstream process steps.[14,15]

The state-of-the-art technology is a two-stage batch-based mixing process.[16] The slurry ingredients are combined into the large container and then stirred until homogeneity is achieved. And during this time, the ability to monitor the potential resulting slurry is limited because the components are constantly undergoing the mixing process. If the electrode slurry is poorly mixed, the conductive additive will not disperse well enough.[11] Therefore, batch mixing process is a relatively expensive operation as it takes longer time to complete and need to consider it capacity to process at one time. This is because the mixing time can range widely depending on the chemistry and batch size and not to mention the addition of a degassing step which could consume a couple of hours.[12] Many technologies are proposed as an effective mixer for mixing electrode slurry, such as, magnetic stirring, ball-mill mixer, and ultrasonic mixer. Planetary mixers are usually used for this purpose.[2,10,12,17]

However, a shift from discrete to continuous processing is clearly evident in the equipment and process technology used. Twin-screw extruders (TSE) are particularly suitable for the continuous mixing process, as they offer a number of advantages over batch-based processes. A TSE is comprised of two screws rotating within a cylindrical barrel. A single screw can be made up of multiple screw elements such as conveyors, kneader, backward elements and other special elements.[16] It is a high intensity device which offers a great flexibility due to its modular construction. The resulted slurry rheological properties will be subject to the screw configuration and screw elements. The material is fed into the screws through a hopper and subsequently blended and transported through the screw by its rotary movement.[18] Meanwhile, the material is sheared between the screws and the barrel wall as the screw threads rotate and move the material along. The screw speed, feed rate, and temperatures of various zones of the system can be controlled. It is this ability to control the constant processing that makes battery slurry production via extrusion manufacturing for batteries so promising. TSE enable continuous mixing with a processing time in the order of minutes instead of hours.[19] TSEs have a number of other advantages, including optimized shear rate and higher product consistency. In addition, less material wastage can be achieved after adjustment and continuous mixing offers higher output with lower space requirements.[12] It can therefore be assumed that TSE will be used in the future due to the large number of advantages.

3. State of the Art

There are various studies that deal with the effects of parameters on the product or intermediate product. Either individual processes or cross-process considerations are carried out. The most important studies are presented as follows.

Haghi et al. developed a tailored digitalization concept for electrode manufacturing. A two-step literature-based and expert-based approach was selected to identify parameters. As a result, a literature-based list of parameters in electrode manufacturing and simplified design structure matrix (DSM) were presented. However, the DSM is a symmetrical DSM, so that no direction of influence is mapped. Instead, it shows which parameters have a relationship to other parameters in principle. Furthermore, the focus of the mixing process is on batch-based processing; the continuous mixing of slurry is not considered. To summarize, a first rough overview of the mixing parameters and possible connections is given.[15]

Bockholt did research on formulation techniques for property-optimized lithium-ion battery electrodes. In particular, the effects of the individual ingredients and their quantity shares on the product are studied. It is investigated how product properties change when the amount of individual components are increased or decreased. The effects of parameters on product parameters are analyzed, e.g., how dynamic viscosity is affected when process parameters such as dispersion time are changed. Not only the slurry is considered, but also the electrode itself (e.g., electrode adhesion, conductivity network, and porosity) and the whole cell (reduced cycle stability). In summary, a comprehensive investigation of selected system components and their effects on product performance and quality is carried out. Not only are the interrelationships and influences identified, but the mode of action is also explained. However, this is only done for selected process and product parameters.[20]

Meza et al. developed a simulation to analyze the material behavior inside a TSE. A wide range of process parameters were considered to predict changes in the production quality of battery slurries. An analysis of the effects of the process parameters on the mechanical strain inside an extruder was performed. In particular, the local shear rate for partially and fully filled screw sections and the density distribution were studied. In the article, the effects of the resulting flow profiles generated by different combinations of process parameters, as well as the geometrical features of the screw elements, on the local mechanical stress in the extruder are indicated by analyzing changes in the shear rate distribution. The overall result is that it is now possible to predict slurry profiles and mechanical stress in the process section of a TSE for various process parameter combinations. This helps to set specific slurry properties in a more targeted manner. However, the research was limited to the parameters required for the simulation. A more detailed or comprehensive analysis of all parameters in the continuous mixing process has not yet been performed.[18]

Wenzel et al. focus on the distribution and homogenization of fine particle systems. The focus of that article is on the dry mixing of NMC particles and CB. The results show that mixing characteristics play an important role, despite the intensive loading in an intensive mixer. By systematically processing and analyzing the results, mixing processes have been described in more detail and important relationships have been found. Conclusions can only be drawn from powder conductivity measurements if the mixing process is understood over its entire duration. In addition, this work demonstrates the existence of important correlations between particle characteristics and process and equipment parameters. As the bulk density increases, the conductivity of the mixture decreases. With this research work important basics for the process understanding were created. However, only one mixing process was considered and no adaptation to other mixing and dispersing processes was made. The continuous mixing process was also not considered.[21]

Dreger et al. examine diverse dispersing devices, both discontinuous and continuous, to assess their effects on the structural and electrochemical traits of electrodes created from commercial NMC cathode material. A laboratory-scale dispersing device was evaluated alongside a discontinuous laboratory kneader and a continuous extruder. It was observed that the dispersing technique and duration significantly influenced both structure and electrochemical performance. Although experiments explored various parameter dependencies, a comprehensive delineation of these dependencies or a focused selection of parameters characterizing the mixing process was not conducted.[11]

In the work of Haarmann et al. detailed investigations are carried out on a continuous mixing process for lithium-ion battery electrodes, where cathode electrodes are produced using a co-rotating TSE. Different material compositions and processing parameters such as screw speed are investigated. Processing routes for feeding the binder to the processing zones as a dry powder or pre-solved in a liquid are being investigated. The produced cathode slurries are analyzed for rheological properties and dispersion degree, i.e., mechanical properties, pore size distribution, particle size distribution and characterized for electrical conductivity. Volume flow was found to be the main factor influencing the reduction of residence time in the extruder, while screw speed and screw configuration had only minor effects. The twin screw speed was found to have a direct effect on viscosity, which can be explained by the different shear rates during processing. In fact, there is some dependence on screw speed, formulation, binder formulation, and fill level. This study provides a deep insight into the process-structure-feature relationships of the continuous compounding process, although a focused selection of parameters characterizing the compounding process was not made.[16]

Westermeier et al. considers the entire production chain of lithium-ion cells, mapping cause-and-effect chains up to the quality characteristics of the final product. Methods such as failure mode and effects analysis (FMEA) are used to identify parameters that directly or indirectly influence the quality parameters of the final product. The quality parameter selection method validates, quantifies and extracts the truly quality relevant influence variables through an iteration of theoretical reduction and experimentation. In addition, relevant parameters are selected through an iterative production chain analysis. This is done in combination with Pareto analysis and experimental studies. The focus is on quality parameters and not on traceability parameters. There are thematic overlaps, but it can be assumed that not all parameters which are relevant for traceability, in particular plant parameters, are considered.[22]

Zanotto et al. examine battery cell production and possible data-driven approaches such as digital twins to optimize it. This study identifies a number of gaps between the parameters that can be measured and the parameters that are needed for the development of some of the computational models, such as the material storage conditions of the slurry. At the same time, a large number of parameters are identified for which the direction of influence is partially known. There is no in-depth analysis of the dependencies between the parameters.[23]

Table 1 summarizes the aforementioned studies. It can be seen that no comprehensive studies have been published to date on the influences and dependencies between the parameters in the continuous mixing process. In particular, no assessment has been made of which parameters are relevant for traceability in the continuous mixing process.

4. Traceability Approach in the Continuous Mixing Process

The general and central objective of this research is to design and develop an advanced traceability system for the continuous mixing process in the battery cell production that allows the finest possible granular conclusions to be drawn about the mixing parameters as well as the slurry composition. Therefore, on the one hand a suitable solution for a tracking and tracing in the mixing process and on the other hand a transparent and secure data storage is needed.

A multi-stage approach is chosen to solve the challenge described above. First, the focus is placed on solving the traceability problems associated with the continuous mixing of different materials and the possibility of mixing different batches. This comprehensive analysis, which is content of this article, includes the identification of all parameters inherent to this complex process. Given the large number of parameters, a reasoned selection of the most pertinent ones is conducted, leveraging well-established scientific methodologies. These carefully selected parameters are of key importance, as they serve as the basis for the subsequent implementation of the traceability solution. In addition, possible strategies for product traceability are being explored in depth. Special challenges have to be considered, such as multiple changes of batch structure in the process and minimizing negative effects on the final product and its quality. The solutions considered can be divided into two main categories: Tracer and Tracer-less. Solutions with tracer include matrix codes, tracer particles and electronic microchips. Tracer-less solutions include mathematical and digital models without any physical tracers. Each approach from both categories will undergo a rigorous and comprehensive evaluation based on sound evaluation criteria. This approach ensures that innovative and unconventional methods are not too early discarded, allowing for a holistic and informed decision-making process.

Upon completion of this detailed evaluation phase, a traceability solution will carefully be formulated and seamlessly integrated into a transparent and secure data storage framework. Extensive validation testing will then be conducted to verify the functionality and suitability of the various proposed solutions, including exploitation of the potential level of information content of the traceability solution. In addition, the limitations

Table 1. Overview of the most relevant publications that investigate parameters influences in the mixing process.

References	Application field	Results and findings	Evaluation
[15]	Electrode Manufacturing	• Development of a tailored digitalization concept for electrode manufacturing • Identification of parameters using a two-step literature and expert-based approach • Presentation of a simplified DSM of parameter influence	• Focus on batch-based mixing, no consideration of continuous mixing • Only simplified DSM, no consideration of *para*-meter influence direction
[20]	Lithium-Ion batteries properties	• Investigation of formulation techniques for optimized lithium-ion battery electrodes • Analysis of parameter effects and quantity of ingredients on product parameters	• Focus only on selected system components and their influence on product performance and quality
[18]	Continuous mixing with TSE	• Development of a simulation to analyze material behavior within a TSE • Analysis of process parameter effects on mechanical strain within an extruder • Prediction of slurry profiles and mechanical stress for various process parameter combinations	• Research was limited to the parameters required for the simulation • No comprehensive analysis of parameters in the continuous mixing process
[21]	Homogenization of fine particle system	• Dry mixing of NMC particles and CB • Identification of correlations between particle characteristics and process and equipment parameters • Creation of important foundations for process understanding	• Consideration of only one mixing process, there is no adaptation to other processes such as continuous mixing
[11]	Electrode Manufacturing	• Examination of various dispersing devices' effects on the structural and electrochemical traits of electrodes • Significant influence observed of dispersing technique and duration on structure and electrochemical performance	• No comprehensive delineation of dependencies or characterization of continuous mixing process parameters
[16]	Continuous mixing with TSE	• Investigation of continuous mixing process • Analysis of material compositions and processing parameters' influence on rheological properties and dispersion degree. • Insight into process-structure-feature relationships provided	• Research was limited to selected parameters
[22]	Lithium-Ion Battery Cell Production	• Consideration of entire production chain of lithium-ion cell, mapping cause-and-effect chains up to quality features of final product • Identification of parameters influencing quality parameters of final product. • Focus on quality parameters through methods like FMEA	• Focus on quality relevant parameters and not on parameters for traceability
[23]	Lithium-Ion Battery Cell Production	• Identification of gaps between measurable parameters and those needed for development of digital twins • Partially known direction of influence for a large number of parameters	• Lack of in-depth analysis of dependencies between parameters

of the developed solution will be highlighted and further potential for future research efforts will be identified. This comprehensive approach aims to significantly improve the quality, reliability and traceability of lithium-ion battery cell production.

5. Own Approach and Methodology

As described in the previous section, the first step is to determine the parameters relevant for traceability. In order to cope with the high complexity of electrode manufacturing and especially the continuous mixing process, which can be characterized by a large amount of information, it is necessary to understand, to design and to improve the process chain and the process as a holistic system. For this purpose, an extensive literature research is be carried out. Afterwards a DSM is then built based on the results. The DSM is particularly suitable for this purpose and can be used for complex products and processes. A DSM is represented as a square N × N matrix, mapping the interactions among the set of N system elements, identically labeled and ordered.[24] Pimmler und Eppinger propose a three-stage approach for the development of the DSM:[25] 1) decomposition of the system into elements, 2) documentation of the interactions between the system elements and 3) clustering.

This is followed by the analysis of the DSM. The relevant parameters were weighted based on their active and passive influence strength and divided into three groups using Pareto analysis. Based on the results of the Pareto analysis and interviews with process and machine experts, the parameters that are relevant for a traceability system were finally selected.

6. Results

In the following section, the individual partial results are explained using the procedure described above.

6.1. Literature-Based Approach for the Parameter Identification

In the first step, the system is decomposed into its elements and the framework is defined, only on mixing process step. This work adopted a literature-based approach to avoid subjectivity and include all the relevant parameters in mixing process. In a next step, already known parameters of the mixing process were researched. From the search strategy suggested by Kitchenham et al.[26] the Web of Science and Scopus database were searched. A manual keyword-based search using the keywords and keyword combinations shown in **Table 2** was used. Based on this, a forward and backward search based on publications identified as suitable was conducted. In addition, the "connected papers" website was used to search for related publications that had not previously been considered. Finally, the AI tool "ResearchRabbit" was used to check whether thematic gaps or author clusters had not yet been considered. As it turned out that all of the relevant papers in the subject area had already been included in the previous search strategies, the analysis of the publications could be continued.

More than 200 publications were used as the basis for the identified parameters described in the following. The main focus of comprehensive research was the publication of research results on electrode production, with the emphasis on the mixing process in pilot lines and industrial plants. Hence, the microscale studies in the fields of electrochemistry and material science were not included. In addition, only the open-access publications or journals with access possibility within the Karlsruhe Institute of Technology were reviewed. In a first step, all papers were preselected by their titles and categorized for further consideration. By analyzing the abstracts of the search results, around 109 articles were found that were considered relevant. The research was carried out between December 2022 and October 2023, focusing on publications from the last 24 years (since 1999). A breakdown of the publications by year can be found in **Figure 1**.

As can be seen in Figure 1, the publications considered focus on the period after 2013. On the one hand, this is due to the increasing research activities in this area in recent years, and on the other hand, since around 2016, more and more studies have been carried out on the use of TSE for the continuous mixing of slurry. The identified publications were then classified based on the analyzed aspect, used material, and applied methods.

6.2. Development of a DSM for the Continuous Mixing Process

The DSM is a suitable methodology to identify the interdependencies for complex products and processes. However, the focus here is only on the dependencies between the parameters, which

Table 2. Search strategy for the development of the DSM.

Keywords used in the query	(Slurry OR Electrode OR Anode OR Cathode OR Lithium ion OR Carbon Black OR Binder OR Electrochemical) AND (Product OR Factory OR Manufacturing OR Process OR Mixing OR Dry Mixing OR Wet Mixing OR Extrusion OR Continuous OR Extruder OR Dispersing) AND (Digital OR Data-driven OR data mining OR machine learning OR Simulation OR Experiment OR Analysis OR Quality OR interdependent)
Field of search	Article title, abstract, keywords
Period covered by the publications	1999–October 2023

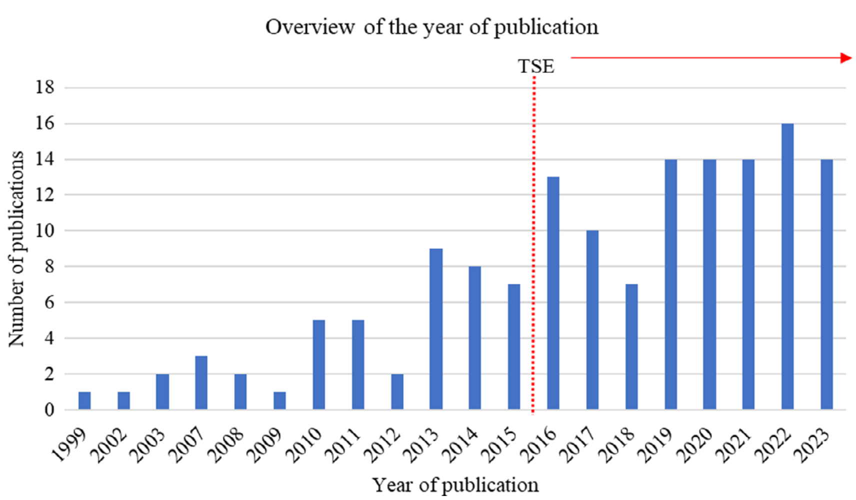

Figure 1. Distribution of the publications considered over the period under review from 1999 to the present day.

indicate the importance of the slurry mixing process in the electrode manufacturing. The aim is to show the dependencies of the parameters within the continuous mixing process based on literature. For this reason, a directed DSM was used, where the dependencies between two parameters are simply represented with an "x" in the matrix. The y-axis has a direct influence on the x-axis and not vice versa. This makes it possible to see which parameter has a particularly large influence on other parameters and thus the relevant parameters can be identified for tracking.

The publications found were analyzed and the dependencies described were systematically recorded. The focus lies on the production research and hence the process steps. All parameters mentioned in the publications were extracted and combined into a complete parameter list. To ensure a consistent presentation and naming, parameters with different names but the same meaning were combined into a single parameter. Based on this list, the parameters were categorized into overarching, raw materials, dry mixing and dispersing according to Kampker et al.[9]

6.2.1. Overarching (Production Environment)

To ensure a good quality of electrochemical performance of the battery cell, it is subject to a great extent on its materials, structure, design and the production environment. Production environment plays a vital role in term of the type of the environment, temperature, atmospheric humidity and pressure along the manufacturing process. Such factors have an impact on the slurry quality that will noticeably at drying and calendaring process.

6.2.2. Raw Materials

The first component of the slurry is the active material. A relevant parameter is the type of active material, such as lithium nickel cobalt manganese oxide (NMC), particle properties, and particle morphology. Particle properties which include particle size of conductive agent comprising their distribution and particle shape that is pivotal in electrode performance.

Apart from that, the particle morphology of the raw materials is a key parameter that can be modified by mixing and dispersing.

Besides active material, binder is the next raw material playing a crucial role in determining the electrochemical performance of the lithium-ion battery. The binder interconnects the active material and the conductive additive and adheres the electrode slurry to the current collector, preventing electrode delamination during the battery cycling procedure.[27] The characteristics of binder need a comprehensive review before it is selected to use. In addition, the analyzed properties of binder for selection are the molecular weight, molar volume, density and the type of material. Among these properties, the chemical stability of the binder is considered the most crucial for its application in the battery cell.

While active materials serve as a reservoir for lithium, the conductive additives or agents are used to increase the electrical conductivity of the slurry. Specific surface area and density should be considered for the material's electrical conductivity. In addition, the particle size is an additional relevant material property.[28]

The solvent is the last component in solvent-based electrode manufacturing to obtain a viscous slurry. The solvent concentration impacts the uniformity and stability of the dispersion and, consequently, the processability of the slurry. The most important properties to be considered for choosing the solvent are viscosity, evaporation rate and boiling point, the solubility of polymers, dispersion stability, surface tension, and flashpoint.[29]

6.2.3. Dry Mixing and Dispersing

The mixing process is a predominant step in electrode manufacturing, having irreversible impacts not only on the electrochemical performance of the battery cell but also on the subsequent process steps. The slurry as a suspension consists of various components differing in size, shape, and density. Eventually, along mixing process, there are challenges regarding the slurry's stability, sedimentation of the large particles, and agglomeration of the small particles. A few parameters that impacting the next manufacturing process are the slurry's processability, uniformity, and stability.[30]

The process parameters in the mixing process include the rotational speed of the agitator, the circumferential velocity, mixing time for suspension production, mixing time for solid powders (dry mixing time), degassing time,[31] cooling temperature of the container during dispersing, applied pressure during dispersing[32] and pressure during degassing.[33] The mixing sequence is identified as an additional aspect influencing the characteristics of the battery cell.[34]

6.3. Analysis of the DSM to Identify the Dependencies between the Identified Parameters and Their Directions of Influence

The following chapter takes a look at the DSM. The influence/impact of a parameter on other parameters is represented by an "x" in the corresponding field. No entry is possible on the diagonal because a parameter cannot influence itself.

Based on the literature reviewed, 61 parameters were identified for which relationships were reported in the publications. These parameters were listed on both the x-axis and the y-axis. An excerpt from the DSM is shown in **Figure 2**. During the research, it became apparent that a simplified DSM, in which only a relationship between the parameters without specifying the direction of the relationship, is unsuitable. Therefore, this DSM is an asymmetric DSM from which the direction of influence can be read. Because it only indicates whether there is an influence or not, no quantitative statement about the influence is possible. It is much more a qualitative statement.

In principle, the DSM shows three different possibilities of influence: 1) The first possibility is that a parameter impacts one or more parameters. The parameter itself is not affected. This is particularly the case with time parameters and sequence parameters (e.g., mixing sequence). This also applies to parameters that describe the energy input into the system. Quantities that depend directly on the selected slurry recipe, such as the amount of binder, amount of active material or solvent, also influence other parameters without being affected by them. 2) The second possibility is that parameters influence one or more parameters and are themselves influenced by one or

Parameter	Type of the environment	Enviroment temperature	Atmospheric humidity and pressure	Rotational speed of the agitator (e.g. screw)	Circumferential velocity	Pressure during dispersing	Specific energy input	Cooling temperature	Process wet mixing time	Dry mixing time	Degassing time	Degassing pressure	Mixing sequence	Geometry of agitator	Filling degree of mixer, e.g. screw barrel	Size of the mixing container	Motor current	Shear stress	Amount of active material	Amount of conductive additives	Amount of binder	...	Solid content
Type of the environment																							
Enviroment temperature																							
Atmospheric humidity and pressure																							
Rotational speed of the agitator (e.g. screw)					x				x						x			x					
Circumferential velocity																		x					
Pressure during dispersing																							
Specific energy input				x	x				x	x								x					
Cooling temperature																							
Process wet mixing time																							
Dry mixing time									x														
Degassing time																							
Degassing pressure																							
Mixing sequence									x														
Geometry of agitator					x				x	x								x					
Filling degree of mixer, e.g. screw barrel									x									x					
Size of the mixing container									x					x				x					
Motor current				x						x					x								
Shear stress																							
Amount of active material																				x	x		x
Amount of conductive additives																			x				x
Amount of binder																			x				x
⋮																							
Solid content										x								x	x	x	x		

Figure 2. Excerpt from the DSM to show the dependencies between the identified parameters and their directions of influence. The corresponding references are noted as comments in the boxes; this is visualized by the small red triangle in the top right-hand corner.[10–13,15,16,18–21,30–168]

more parameters. These are often process and product parameters. An example of this is the homogeneity of the slurry, which influences the dynamic viscosity, sedimentation rate and slurry stability, for example. Conversely, the homogeneity of the slurry is influenced by a variety of parameters such as the mixing sequence and mixing time, as well as the material proportions. 3) The last possibility is that a parameter does not affect other parameters, but is itself influenced. A typical example of this is product parameters such as the electrochemical performance of the slurry and the conductivity of the slurry.

Based on the DSM and the impact direction of parameters, an analysis of the influence strength of each parameter is possible. The aim here is to find out which parameters have a particularly strong influence on other parameters or are influenced by a particularly large number of parameters, the so-called active and passive sums. The active sum is the number of influences of a parameter on other parameters. This involves recording how many parameters are influenced by a single parameter. With the passive sum, the reverse scheme applies, i.e., how many parameters influence the parameter under consideration. To determine the influence strength, the number of influences per line is summed up to the active sum and then set in relation to the total sum of all lines. This is the active influence strength. For example, the parameter "solid content" influences 20 other parameters (= active sum). In relation to the total sum of 206 influences shown in the DSM, this results in an active influence strength of 9.7% for "solid content". **Figure 3** shows the active influence strength of parameters and **Figure 4** the passive influence strength of parameters.

Figure 3 shows which parameters have a major influence on other parameters. For this purpose, the number of × in the DSM was summed up in the x-direction. The proportion of the total was then calculated, e.g., almost 10% of all active influences can be attributed to the solid content. In the graph above, this is visualized using blue columns and the parameters are listed along the horizontal axis in descending order of influence strength. The orange line represents the total sum of influence of several parameters. For example, 40% of all active influences are caused by the parameters solid content, rotational screw speed, specific energy input, amount of binder, conductive agent distribution, dynamic viscosity and yield point of slurry.

The same calculation method was used for the passive sum and passive influence strength. Here the influences of the individual columns (y-direction) are calculated and set in relation to the total sum. The example of the parameter "particle size distribution" shows that this parameter is influenced by 13 other parameters (= passive sum), resulting in a passive influence

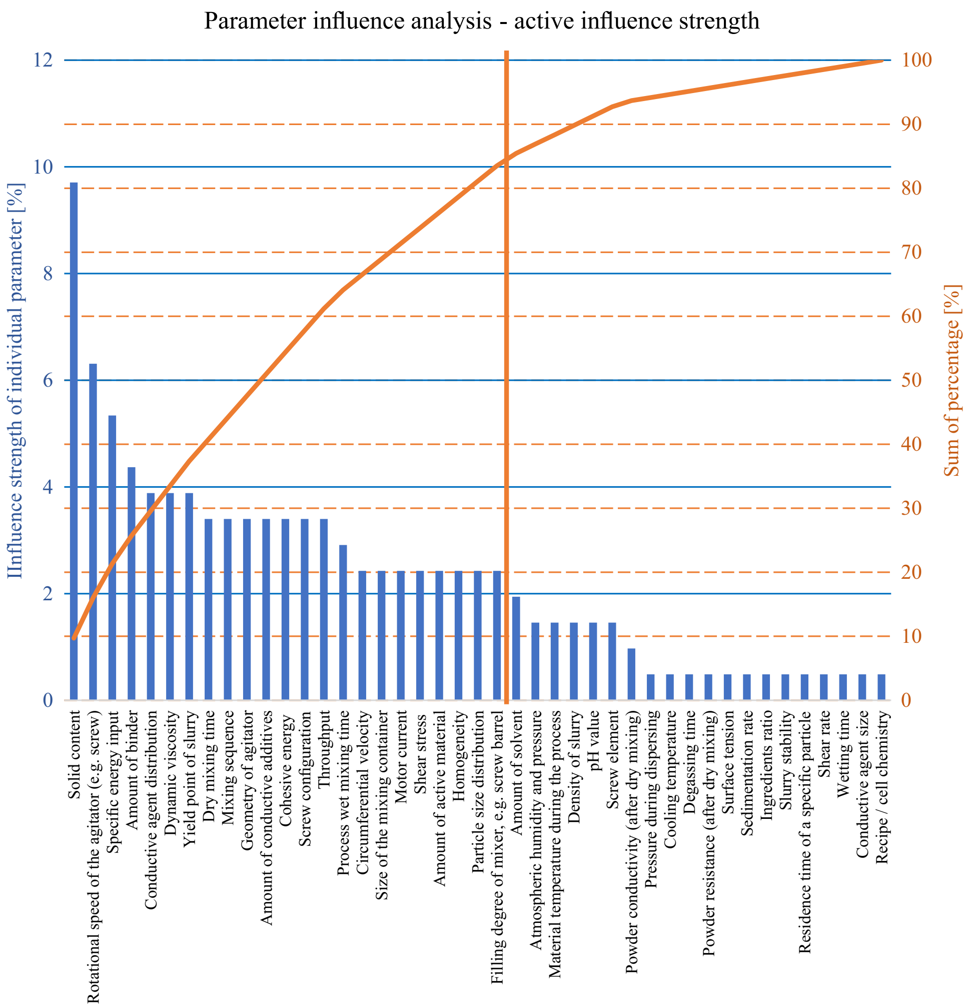

Figure 3. Analysis of the active parameter influence (active influence strength).

strength of 6.3%. The expected degressive curve of the orange curve can also be seen in Figure 4.

It is clearly recognizable that the dynamic viscosity is most strongly influenced by other parameters. Together with the dynamic viscosity parameter, the parameters sedimentation rate, slurry homogeneity, particle size distribution and shear stress in process account for a total of 43% of the total influence (passive influence strength).

Based on the active and passive sums, the total influence strength of the individual parameters was calculated. It can also be derived from this which parameter has a major influence.

A Pareto analysis was used to determine the proportion of influencing variables or factors in the overall effect on a result variable. The parameters were divided into three groups: "major influence" (80%), "minor influence" (15%) and "negligible influence" (5%). Based on this grouping, the parameters that were assigned to the group with a major influence were considered to be particularly relevant. When classifying the groups, however, it should be kept in mind that a strict classification based on the percentages mentioned is not expedient. For example, it could be that a parameter is still counted in the group with "major influence" and a parameter with the same relative influence is counted as "minor influence". With regard to the parameters of the active influence strength, this means that although the "filling degree" parameter has the same influencing power as the "shear stress" parameter, the total sum is already 81,1% before this parameter. According to a strict interpretation of the Pareto rule, the parameter would therefore no longer be considered, although the influence of the filling degree on other parameters is comparable to that of shear stress. Therefore, the groups were divided in such a way that all parameters with the same relative influence were always assigned to the same group. Specifically, this means that all parameters up to and including filling degree of mixer (83,5% active influence strength) in Figure 3 and all parameters including filling degree of mixer (80,6% passive influence strength) in Figure 4 were assigned to the "major influence" group. The result is the list in **Table 3**. This shows all 30 parameters that have a particularly large influence.

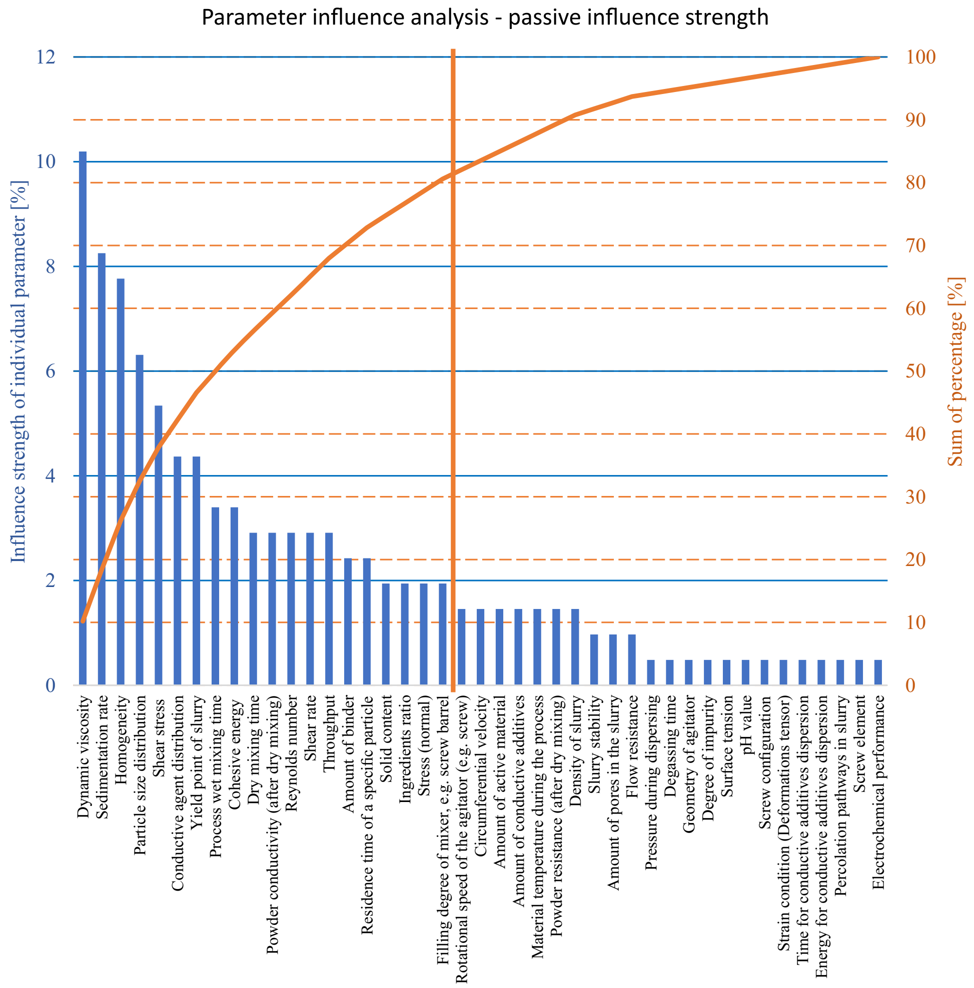

Figure 4. Analysis of the passive parameter influence (passive influence strength).

Table 3. List of parameters with the greatest influence.

No.	Parameter	Share of total influence	No.	Parameter	Share of total influence
1	Dynamic viscosity	14,1%	16	Filling degree of mixer, e.g., screw barrel	4,4%
2	Solid content	11,7%	17	Amount of conductive additives	3,4%
3	Homogeneity	10,2%	18	Mixing sequence	3,4%
4	Particle size distribution	8,7%	19	Geometry of agitator	3,4%
5	Sedimentation rate	8,3%	20	Screw configuration	3,4%
6	Conductive agent distribution	8,3%	21	Powder conductivity	2,9%
7	Yield point of slurry	8,3%	22	Reynolds number	2,9%
8	Shear stress	7,8%	23	Shear rate	2,9%
9	Cohesive Energy	6,8%	24	Circumferential velocity	2,4%
10	Amount of binder	6,8%	25	Amount of active material	2,4%
11	Rotational speed of the agitator	6,3%	26	Residence time of a specific particle	2,4%
12	Process wet mixing time	6,3%	27	Size of mixing container	2,4%
13	Dry mixing time	6,3%	28	Motor current	2,4%
14	Throughput	6,3%	29	Ingredient's ratio	1,9%
15	Specific energy input	5,3%	30	Stress (normal)	1,9%

7. Discussion and Determination of Relevant Parameters for Traceability

In the analysis carried out, the authors assumed that the parameters with the greatest active influence are not the parameters that are most strongly influenced. It was expected that the parameters specified by the plant operator would predominantly have a significant influence on other parameters, especially those directly related to product specifications. A comparison of Figure 3 and 4 confirms this expectation. As the two graphs and Table 3 show, the seven parameters with the highest influence values are all related to product properties and together account for over two-thirds of the total influence.

It was not surprising that dynamic viscosity had the highest passive influence strength. It should be noted that the distribution of the conductive agent also has a very high influence strength. A possible explanation could be that this parameter has a profound influence on the structure of the slurry and thus on the coated electrode, and therefore also on other parameters.

In addition, the authors would like to point out that, according to the analyses carried out, the screw speed has a greater influence than the screw configuration of the twin screw extruder. This was not in line with expectations, as it was expected that the screw configuration would have a greater influence value due to its significant influence on parameters such as shear stress, specific energy input, shear rate and residence time of certain particles. A possible explanation offered by the authors is that the screw configuration has an indirect effect on many of the parameters and a direct effect on only a few parameters. Due to the tendency of the studies considered to consider mainly direct influences in their investigations, the chosen methodology consequently results in lower influence values than would be expected based on the experience of the authors and industry experts. In addition, the studies reviewed primarily varied the speed of the mixer and rarely compared different mixer or screw geometries. This may also have had an impact on the results of the analysis.

It should also be noted that the residence time of certain particles has a lower influence value than expected. However, the authors emphasize that its influence value is comparable to other relevant parameters, such as the amount of active material, and almost equivalent to the shear rate parameter. This underlines its importance.

Overall, the analysis is consistent with the authors' own experience and that of industry experts, confirming the validity of the results.

The dependencies identified in this publication are derived from extensive experimental studies by various authors. Therefore, the comprehensive summary presented here represents an aggregation of effects that have already been identified and demonstrated. However, it should be noted that this publication only includes effects that have already been documented in the existing scientific, research and industrial literature. It is possible that there are unknown effects and interactions that have not yet been researched or documented. It is also possible that effects and influences of parameters have not been considered by other researchers because they were not in focus, the measured values were too small and may have been interpreted as a noise signal, or were considered insignificant and therefore not published.

For this reason, additional experimental validation is considered beneficial, especially in conjunction with a comprehensive design of experiments (DoE). However, a well-structured and statistically validated experimental design is essential. It is important to consider that conducting experimental studies will require a significant amount of effort, and therefore, the cost-benefit ratio should be carefully evaluated.

In the studies under review, the preparation of the pre-mix and powder mixtures is not described or is inadequately described. Therefore, this aspect of pre-mixing was not further considered in this publication and the effects of different pre-mixes on the mixing process and parameter influences were not further investigated. However, it is important to mention that the pretreatment of materials can have a significant impact on the final slurry properties. Therefore, the authors recommend that the effects of pre-mixes be considered in the context of experimental testing and validation of the results presented here.

Based on the discussed results and the resulting findings, the parameters relevant for traceability in the continuous mixing process are now determined. It was found that product parameters have a high passive influence strength. In addition to the product parameters, consideration of the process and plant parameters is relevant for a holistic traceability system. It should be possible to characterize the mixing process using the parameters relevant for traceability. When recording the parameters that characterize the mixing process, in-line and on-line recording is particularly preferable in order to be able to detect and react to changes in the process and product at an early stage.

As shown by the parameters listed in Table 3, it becomes clear that some parameters cannot be recorded in-line, but only at-line or off-line. For example, it is not yet possible to detect the shear rate, shear stress or specific energy input in-line in the continuous mixing process. The same applies to the sedimentation rate. A particular challenge here is the determination of the residence time of a specific particle, for which the authors are not yet aware of any in-line method in the field of battery production. Other parameters such as the homogeneity of the slurry can only be indirectly recorded or estimated in-line.

This limitation means that not all of the parameters that have a major influence or are strongly influenced and thus characterize the process can be continuously recorded and are suitable for the development of a traceability system. A selection must therefore be made from these parameters.

The selection of parameters for a traceability system requires a criteria-based approach. The parameters to be considered must be included in the list in Table 3 and therefore have a high influence value. It is important to consider both product and process parameters. Process parameters include machine components that interact directly with the product, such as screw configuration. It should also be noted that the selected parameters should be measurable using currently known and commercially available measurement methods. While certain parameters cannot be measured directly, they can be determined analytically from other parameters using a "soft sensor approach". Therefore, these parameters will not be given primary consideration in this publication and will not be included in the list of parameters to be tracked. Another important constraint is that the selected

Table 4. List of product and process parameters to be tracked that characterize the continuous mixing process and should therefore be tracked.

No.	Product parameter	No.	Process parameter
1	Amount of active material	1	Screw configuration
2	Amount of conductive additives	2	Mixing sequence
3	Amount of binder	3	Rotational speed of the agitator
4	Amount of solvent	4	Geometry of agitator
5	Ingredient's ratio	5	Motor current
6	Dynamic viscosity	6	Throughput
7	Solid content	7	Filling degree of mixer
8	Particle size distribution	8	Volume flow rate
9	Residence time of a specific particle	9	Dry mixing time
10	Material temperature during process	10	Process wet mixing time
11	Density of slurry	11	Size of mixing container
12	Powder material conductivity	12	Actual temperature in the mixing section
13	Homogeneity	13	Specific energy input
14	Conductive agent distribution	14	Residence time distribution

parameters should ideally be automatically measured in-line or on-line. Although parameters that can only be determined off-line in the laboratory are potentially valuable to a traceability system, in most cases they cannot be integrated into a continuous and automated traceability system. Therefore, for the time being, these parameters will not be given primary consideration in this publication and will not be included in the list of parameters to be tracked. They can therefore be included in the list of tracked parameters.

The parameters that, according to DSM and Pareto analysis, have the greatest influence on the continuous mixing process and meet the above criteria are listed in **Table 4**. Based on these parameters, a process characterization is possible. The list can and should be adapted and expanded as needed and/or if further findings are gained. The authors recommend to consider the constantly growing expertise of the industry and, if necessary, to add new parameters in the parameter list based on this knowledge.

A very large proportion of the parameters that need to be recorded can already be recorded using implemented system technology and sensors. However, the corresponding sensors still need to be retrofitted, especially for an exact determination of the flow rate. Furthermore, an appropriate measurement setup in the form of a single or several sensors must be integrated to determine the residence time distribution. Depending on the "tracer particle" used, a conductivity sensor, a camera or an NIR sensor could be used. The sensors must have the appropriate sensitivity and repeatability.

8. Conclusion

This study addresses the critical issue of traceability within the mixing process. The issue of traceability within the mixing process is of importance due to its role in maintaining quality and ensuring product consistency. To address this challenge, a comprehensive procedure and concept were introduced. Extensive research and analysis of the literature was carried out for this purpose. Based on this knowledge, the parameters and their interdependencies were identified. This included the creation of a DSM to examine parameter influences in the continuous process and identify crucial parameters. The developed DSM was examined using Pareto analysis and the parameters with the greatest influences were identified. It turns out that a few parameters have a particularly large influence. For example, the solid content, rotational speed of agitator and specific energy input have a particularly strong influence. Other parameters such as dynamic viscosity, sedimentation rate and homogeneity of the slurry are influenced by a large number of parameters. Based on this and expanded by expert interviews, a list of parameters was created that are essential for effective traceability in the continuous mixing process.

Now that it is known which parameters should be covered by a traceability system, the authors plan to take a closer look at possible solutions for traceability in the continuous mixing process as a next step. For this purpose, a morphological box will be developed to investigate possible solutions without tracers such as residence time distribution and particle-based solutions such as RFID chips. Based on the results, an in-depth analysis of the most promising solution will then be considered and a complete traceability system will be developed.

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Conflict of Interest

The authors declare no conflict of interest.

Author Contributions

S.O.: Conceptualization; S.O.: Methodology; S.O.: Formal Analysis; S.O. and N.N.A.M.S: Investigation; S.O.: Visualization; S.O.: Validation; S.O. and N.N.A.M.S.: Writing—original draft preparation; S.O., S.S. and J.F.: Writing—review and editing; J.F.: Supervision & Funding. All authors have read and agreed to the published version of the manuscript.

Data Availability Statement

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

Keywords

battery cell production, continuous process, design structure matrix, mixing, parameter interdependencies, slurry, traceability

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