

# Targeted data generation in the continuous production of anode slurries for lithium-ion battery cells

1<sup>st</sup> Sven Oberdiek

*Quality and reliability management  
Fraunhofer Institute for Manufacturing  
Engineering and Automation IPA  
Stuttgart, Germany  
sven.oberdiek@ipa.fraunhofer.de*

2<sup>nd</sup> Leah Jalowy

*Dispersing Technology  
Fraunhofer Institute for Manufacturing  
Engineering and Automation IPA  
Stuttgart, Germany  
leah.jalowy@ipa.fraunhofer.de*

3<sup>rd</sup> Flavio Gonzalez Vazquez

*Purity-specific automation systems  
Fraunhofer Institute for Manufacturing  
Engineering and Automation IPA  
Stuttgart, Germany  
flavio.gonzalez.vazquez@ipa.fraunhofer.de*

4<sup>th</sup> Monika Risling

*Data and application services for  
digital production  
Fraunhofer Institute for Manufacturing  
Engineering and Automation IPA  
Stuttgart, Germany  
monika.risling@ipa.fraunhofer.de*

5<sup>th</sup> Katja Wahl

*Purity-specific automation systems  
Fraunhofer Institute for Manufacturing  
Engineering and Automation IPA  
Stuttgart, Germany  
katja.wahl@ipa.fraunhofer.de*

**Abstract**—This research paper explores the transition from batch mixing to continuous mixing processes for the production of slurries used in lithium-ion battery cells. The conventional batch mixing methods, prevalent in European industry, suffer from time-consuming cleaning processes, lengthy mixing times, and the inability to monitor paste conditions in real-time. The study proposes a solution using continuous mixing with a twin-screw extruder, highlighting advantages such as reduced cleaning time, immediate sample characterization, and minimized production risks. The research aims to develop a test setup and procedure to accelerate the understanding of the continuous mixing process, leveraging minimal resources and time, and facilitating the training of artificial intelligence algorithms. The ultimate goal is to create a digital twin encompassing all influencing factors for predictive and prescriptive analytics. The methodology involves literature reviews, expert surveys, and experimental setups, with an emphasis on inline measuring devices. The paper presents results related to relevant product characteristics, influencing factors, and the selection of suitable measuring equipment. The experimental setup includes a database structure, human-machine interface, and visualization of measurement data. The study concludes with insights into the correlation analysis of influencing factors and product characteristics, emphasizing the need for further experiments and the development of algorithms for predictive quality assessments in continuous mixing processes.

**Keywords**—*lithium-ion battery slurries, continuous mixing, twin-screw extruder, batch mixing, data collection, process know-how, inline measuring, predictive quality, artificial intelligence, digital twin*

## I. INTRODUCTION

The production of slurries for lithium-ion battery cells is primarily carried out in batch mixing processes, especially in the European industry [2]. The pastes are produced in a multi-stage process in the plants, which often have a capacity of up to 3,000 liters. Very time-consuming cleaning processes are sometimes necessary between the individual process steps.

The actual mixing process is also time-consuming and often takes several hours [3, 4]. According to the current state of the art, it is not possible to monitor the condition of the paste during the mixing process, which is why offline material characterization is necessary afterwards. Only after successful characterization is the paste released for further production, i.e. the coating of the electrode sheets. This lengthy production process limits the maximum production output. Increasing electrification in all areas of life is also leading to rising demand for lithium-ion batteries [5]. In order to meet the increasing demand, appropriate technologies with higher output quantities are required. A challenge connected to this is producing consistent quality across multiple batches [3, 6].

One possible solution is continuous mixing with a twin-screw extruder [2, 7, 8]. The material is fed into the extruder via various connections and automatic dosing systems. The position of the feeders can be varied as required. Mixing takes place in a chamber with two either counter-rotating or co-rotating screws [9]. Continuous mixing is characterized by various advantages over batch mixing. For example, there is no need for time-consuming cleaning of the mixing tools and containers as long as the same recipe is produced over a relatively long period of time. In addition, an initial material sample can be taken promptly after the start of the process to characterize the slurries. Therefore it is possible to determine the produced quality much earlier. This reduces the risk of rejects compared to batch mixing. The continuous mixing process is particularly suitable when large quantities of a recipe are produced and no material changes are made [10]. However, running in the process is complex due to the large number of influencing variables [11].

Batch mixing allows to test the batch once for its characteristics and therefore determine the quality of the entire batch. Continuous mixing requires permanent testing in order to prevent quality issues in later production steps. For constant monitoring of the production quality, the reduction of the processes complexity and to improve the knowledge of

coherences data driven models like digital twins, data analytics and predictive analytics offer a solution.

## II. MOTIVATION

A technology change from batch mixing to continuous mixing poses challenges for companies. For example, it can be assumed that extensive process know-how is not yet available at the time of the changeover. In addition, the literature provides only a few preliminary studies that were primarily carried out under laboratory conditions, and these mostly relate to cell performance and not to the process know-how required for electrode production [12–14]. This work addresses this problem. The development of process know-how is usually associated with a lot of time and effort. In addition, no data-driven analysis methods can be used to extract knowledge, as there is simply no or very little data available about the new processes. Therefore, the aim of this work is to develop a test setup and procedure that makes it possible to build up the deepest possible understanding of the process with minimal resources and time. The research question of this work is how a testing setup for data generation in the continuous mixing of battery slurries must be designed to allow quick results considering the gain of process knowledge and the training of artificial intelligence algorithms.

The long-term goal is to create a digital twin of all influencing factors and product characteristics of the process of continuous mixing of battery slurries, which can be used for predictive and prescriptive analytics.

## III. RESEARCH METHODOLOGY

The methodological approach of this research consists of three overarching steps. Firstly, the experiments and data collection are prepared. The preparation is based on a literature review of previous work in the field of cause-and-effect relationships in the continuous mixing of slurries for lithium-ion battery cells and an expert survey. The survey is conducted with experts in battery cell production, dispersion technology and extrusion technology. This process step aims to identify possible influencing factors and product characteristics to be manufactured and to qualitatively evaluate the influence of the individual influencing factors on the product characteristics.

The measurement and data concept is then designed on the basis of this evaluation. Additional input for this step is provided by the requirements with regard to the digital technology connection, the requirements for the measurement data to be collected and the expectations of the machine operators with regard to the graphical user interface (GUI). The results of this step are an initial prototype of the GUI, the selection of suitable measuring devices and a database structure including a data connection plan.

The final step is the testing and verification of the test setup. The input is a two-factor test plan previously developed on the basis of the influencing factors and measured values from samples characterized offline. The aim of this step is to create a preliminary database and verify the developed test setup in order to subsequently carry out further tests and thus generate a more meaningful database. Figure 1 shows the methodical procedure including input and output of the respective method step.

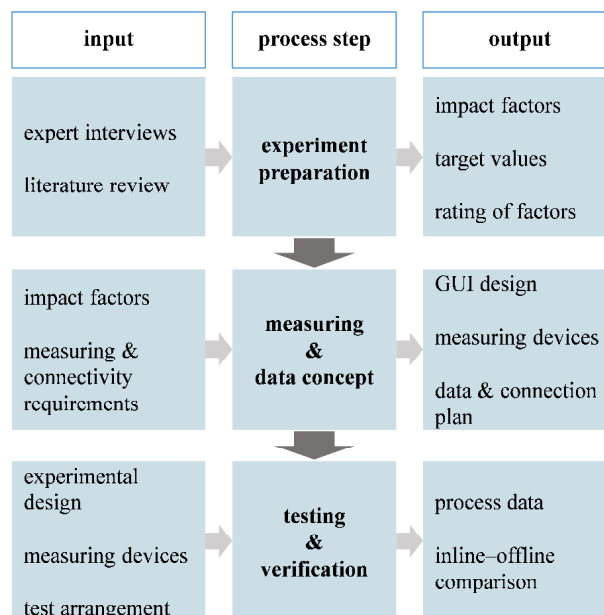


Fig. 1. Process of the applied method

As organic solvents are used for the production of cathode material and therefore, special occupational safety regulations apply and special testing equipment is needed, this paper only considers anode material [15]. Different formulations are also not considered.

As presented in the previous chapter, literature research together with expert interviews and expert workshops form the basis for the further procedure in this thesis. The aim is to collect potential influencing factors of the production process and product characteristics to be manufactured. This is followed by a qualitative evaluation of each influencing factor in relation to the various product characteristics.

The evaluation is based on values from 0 to 3, with 0 indicating no influence of the factor on the product characteristic and 3 indicating a high influence. It is also possible not to evaluate a factor/characteristic combination if the state of scientific knowledge does not allow a statement to be made. Unevaluated influences are indicated by a minus sign. Figure 2 shows a schematic representation of the qualitative evaluation matrix.

evaluation matrix		influence factors		
		factor 1	factor 2	factor 3
product characteristics	characteristic 1	1	0	2
	characteristic 2	1	2	3
	characteristic 3	2	3	1

Fig. 2. Template of the used evaluation matrix

The evaluation matrix can be used to prioritize influencing factors and product characteristics. The prioritization allows targeted preliminary tests to be carried out. In addition, the target values of the respective product characteristics and the associated tolerances are recorded. This information forms the basis for the subsequent selection of suitable measuring equipment.

#### IV. RESEARCH RESULTS

Expert interviews and research have shown that viscosity, density, particle size distribution and temperature are relevant product characteristics. Viscosity is particularly relevant against the background of further processing during coating, while the particle size distribution provides information on homogeneous mixing of the slurry [16]. Therefore, measuring devices for density, particle size distribution and viscosity are required to characterize the slurry. In addition, the temperature of the paste must be monitored, as the measured density and viscosity are temperature-dependent [16, 17].

The influencing factors determined and the sum of their weighting across all product characteristics, in parentheses, are the speed of the extruder's main drive (6), the throughput (5), the setting of the vacuum valve (3), the system temperature control (5), the position of the liquid dosing (6) and the configuration of the screws (4).

As changing the screw configuration and changing the positioning of the liquid dosing unit in industrial systems involves considerable additional work, these factors are no longer considered below. In addition, the degree of homogenization of the paste produced is not considered in this work. This is planned for future tests.

The selection of suitable measuring equipment is limited to inline measuring devices, as these allow the paste to be characterized immediately after production. This minimizes the time lag between production and characterization. Inline measuring devices also make it possible to avoid influencing factors such as inaccuracies during sampling and characterization, for example, due to excessive time lags between sampling and testing. In addition, continuous sampling of the slurries produced is possible, which makes the effects of process fluctuations on the product characteristics visible. The requirements for the measuring devices also include a standardized process interface, a uniform and standardized digital communication interface where possible and coverage of the required measuring range.

The following table lists the measuring devices used and their respective properties.

TABLE I. Criteria for the selection of measuring devices and their properties

criteria	density	viscosity	particle size distribution
Measuring range	< 3,000 kg/m <sup>3</sup>	250 mPas – 12,500 mPas	0.1 μm - 3,000 μm
Process connection	tri-clamp	tri-clamp	tri-clamp
digital connection	ModbusTCP	ModbusTCP	TCP/IP

The data recorded by the measuring devices during the test was merged with the machine control data in an open source time series database. The database is connected to a human-machine interface (HMI) on which the measurement data is displayed visually using open source software. The schematic representation of the test setup is shown in Figure 3. This setup makes it possible to monitor the process during the test and draw initial conclusions about the functionality of the system.

Recording the influencing variables also enables precise planning of the test procedure. With the help of factorial test

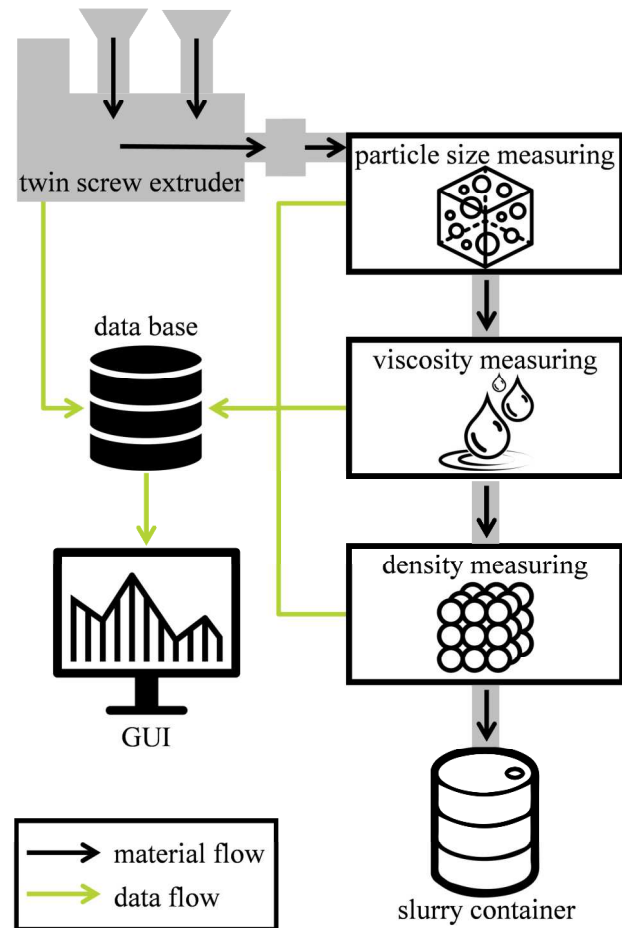


Fig. 3. Material and data flow of the test setup

planning, the number of tests to be carried out can be reduced compared to the one-factor-at-a-time method. A test plan with two stages and five factors results in 32 individual tests [18]. Since parameters on the extruder change after each test and a certain process time elapses until all material of the old parameters has passed through the test setup, this means a high expenditure of time and material. This is not an expedient procedure for an initial preliminary examination of the test setup. For this reason, a reduced analysis is initially carried out using a screening test plan of resolution V, which offers a low risk of misinterpretation of the results, as all two-factor interactions can be considered. The number of tests to be carried out can thus be reduced from 32 to 16 [18].

Carrying out the tests and cleansing the data generates the data volumes per product characteristic shown in the following table. Due to technical issues with the testing devices and the twin-screw extruder during several test runs the amount of usable data is highly reduced. Only data from 6 out of 16 tests are included in the following.

Table II. Amount of collected data per property after cleansing

measured property	density	viscosity	particle size distribution
Amount of data entries after cleansing	6,120	5,037	401

The further evaluations are limited to the viscosity, as no further results were available at the time of writing. The more detailed analysis of the available measured values does not allow any conclusions to be drawn about significant correlations between the respective influencing factors and the viscosity. The correlation is -0.375 for the vacuum valve setting, 0.375 for the speed, 0.257 for the throughput and 0.385 for the system temperature control. Over all carried out tests the inline measure viscosity varied between 670 mPas to 787 mPas with a mean absolute deviation from the arithmetic mean of 24 mPas. This relatively low deviation also shows the minor impact of the conducted parameter changes in the individual test runs.

In addition to the pure data recording, the inline measured values were compared with offline measured values. For the individual tests, additional manual samples were taken, the viscosity of which was determined on a rheometer. While the viscosity measured inline averages 670 mPas, the viscosity measured offline averages approx. 880 mPas. The difference between the measured values can be attributed to the different measurement principles of offline and online measurement. This can be dealt with by determining an offset. However, this requires further testing. Based on the available data, no clear rule can be derived for the comparability of the data from inline and offline measurements.

## V. CONCLUSION AND OUTLOOK

The results presented show how an experimental setup can be designed for targeted data generation in the continuous mixing of anode slurries for lithium-ion battery cells. The basic functionality of the experimental setup has also been demonstrated on the basis of the data. The methodical approach of this work can be used to create a low-cost database for process analyses and to generate process know-how. This work thus lays the foundation for creating a more comprehensive database in further tests, which includes factors not considered in this work, such as the positioning of the liquid dosing and longer test run times. In addition, the degree of homogenization can be considered in the future. AI

algorithms can be trained with the help of such a database. Following this work, a Bayesian network is first modeled, as this allows both qualitative and quantitative data to be correlated. In addition, correlations between the individual influencing factors can be visualized in this way [19]. Based on this, individual influencing factors are examined in more detail and supplemented with quantitative data. The aim is to gradually map all relevant influencing factors of extruder mixing digitally. In future, for example, the inline measuring devices could be replaced by predictive quality algorithms. There is a need for further research into the development of algorithms for determining parameters for fluctuating process input variables such as batch fluctuations in the raw materials. In summary, it can be said that this work answers the question of how an experimental setup can be designed with the aim of generating data which in a later step can be used to train predictive and prescriptive algorithms for continuous mixing. However, further experiments are required to make valid statements on correlations between the influencing factors and product characteristics as well as interactions between them. Also further data are needed in order to train above mentioned algorithms.

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