Review

A Systematic Literature Analysis on Electrolyte Filling and Wetting in Lithium-Ion Battery Production

Nicolaj Kaden [1,2,*] **, Ricarda Schlimbach** [3] **, Álvaro Rohde García** [1] **and Klaus Dröder** [1,2]

1. Institute of Machine Tools and Production Technology, Technische Universität Braunschweig, 38106 Braunschweig, Germany
2. Battery LabFactory Braunschweig, Technische Universität Braunschweig, 38106 Braunschweig, Germany
3. Chair of Service Information Systems, Technische Universität Braunschweig, 38106 Braunschweig, Germany
* Correspondence: n.kaden@tu-braunschweig.de; Tel.: +49-531-391-7656

Abstract: Electrolyte filling and wetting is a quality-critical and cost-intensive process step of battery cell production. Due to the importance of this process, a steadily increasing number of publications is emerging for its different influences and factors. We conducted a systematic literature review to identify common parameters that influence wetting behavior in experimental settings, specifically focusing on material, processes, and experimental measurement methods but excluding simulation studies. We reduced the initially found 544 records systematically to 39 fully labeled articles. Our profound analysis guided by attributed labelings revealed current research gaps such as the lack of a holistic view on measurement methods for filling and wetting, underrepresented studies relevant to series production, as well as the negligence of research targeting the transferability of results from the material to the cell level, while also examining the measured variables' interactions. After comparatively illustrating and discussing implications of our findings, we also discussed limitations of our contribution and suggested ideas for potential further research topics.

Keywords: lithium-ion battery; battery production; electrolyte filling; electrolyte wetting; systematic literature review; measurement methods

1. Introduction

Given the irreversible effects on the global climate, there is a collective societal challenge to reduce CO_2 emissions. Research in battery technology has the potential to provide a solution for carbon-neutral mobility by increasing the efficiency as well as expanding the utility of storage options for electrical energy [1]. In this context, high capacity lithium-ion batteries have the potential to transform the mobility industry in the short and medium term by replacing fossil fuels. As a result, the demand for large-format battery cells with high specific capacities and power has increased rapidly in recent years [2]. The implementation of the EU regulation for CO_2 reduction in the automotive sector indicates that this trend will likely continue [3].

Thus, this rising demand necessitates more efficient battery production in order to save time, material, and the associated costs. Battery cost drivers include, in addition to raw material acquisition costs, inefficient manufacturing processes [4]. The process of electrolyte filling, due to long storage times of cells, represents a promising entry point for cost savings [5,6]. This process step is critical for quality, as insufficient wetting of the cell stack materials can result in poor cell performance [7].

Although the potential for savings has been widely recognized in both research and industry, findings covering electrolyte filling remain fragmented. There are complicated interactions between process–structure–characteristics and their relationships, which are often related to prior process steps. Existing research focuses on highly application-specific industrial processes, covers simulation studies without experimental foundation, or investigates isolated aspects such as material or process influences separately. However,

a comprehensive scientific analysis of experimental results on material and process parameters is lacking. Other recently published literature reviews consider the process chain of battery technology, which is related to our focus because of electrolyte filling but shows a gap regarding the focus on future challenges [4]. Material-based topics are again addressed by Zhang in reviews published in 2006 and 2007 but mainly deal with the separators and electrolytes present in the battery [8,9]. Our approach is thus to clearly target experimentally designed lithium-ion batteries and the processes and their experimental measurement methods, whereas we exclude simulation studies to strongly highlight our experimental focus.

In this regard, we intend to contribute by structuring and discussing existing scholarly findings following the approach of a systematic literature review focusing on experimental studies on the topic.

Our article is structured as follows: Section 2 provides the research background on electrolyte filling, wetting, and its anchoring in the process chain, before we expand upon our methodological approach for the systematic literature review in Section 3. In Section 4, we present our findings by reviewing the identified measurement methods on the material and cell levels by retrieving their respective frequency and application. We compare these in illustrative figures as well as tables and reveal research gaps, which are further discussed in Section 5. We also give guidance for potential research topics for future investigations.

2. Research Background

As an introduction, this section explains the research background. First, it provides a detailed explanation of the electrolyte filling process, before its positioning in the context of the overall battery production process chain. Finally, we discuss a plethora of measurement methods used in this process step.

2.1. Explanation of the "Electrolyte Filling" Process

The electrolyte filling process consists of two phases: dispensing (when referring to the first sub-process, we use the rather unusual term "dispersing" rather than "filling" to better differentiate the sub-process from its superior process step of electrolyte filling), also often called filling, and wetting. The general objective of electrolyte filling is to introduce the necessary amount of electrolyte into the cell and ensure that all pores of the cell composite materials are fully wetted and filled. This enables lithium ions to be transported throughout the electrode material, maximizing the battery cell's theoretical capacity [10]. The dispensing sub-process is defined as the period during which the liquid electrolyte is introduced into the dead volume of the cell (cf. Figure 1b). The dispensing process is completed when the cell is sealed.
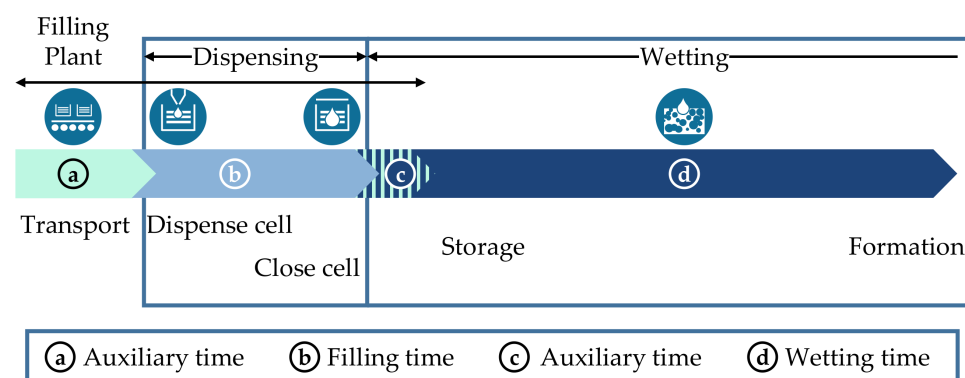

Figure 1. Schematic illustration of the electrolyte filling process with its sub-processes.

Applied research suggests various procedures for determining the amount of electrolyte to be used. These approaches can be divided into different categories:

- Scaling can be direct, based on the pore volume of all cell composites (electrodes and separators) [11].
- Scaling can be indirectly influenced by the separator based on the theoretical capacity of the battery cell, which changes with the area and thickness of the electrodes and therefore correlates with the pore volume of the electrodes.
- Scaling can refer to the weight of the cell composite [12]. The latter changes proportionally to the area and thickness of the electrodes and the separator, resulting in a dispensing quantity that is proportional to the pore volume.
- Moreover, scaling can be based on the electrode surface, where electrochemical reactions between the electrolyte and the cell components occur [13]. These approaches typically rely on the assumption that all pores must be fully wetted.

Wetting, the second sub-process, begins immediately when the cell composite materials come into contact with the electrolyte as it starts flowing into the dead volume of the cell housing (cf. Figure 1d) [14]. In the microporous cell composite material, capillary forces drive the electrolyte to be absorbed into the pore structure, displacing present gas. The residual gas present in the cell can either be found in locally confined accumulations in the material, or it diffuses to the edges of the pore system and is present as bubbles in the electrolyte [15]. If wetting is insufficient, lithium plating may occur during the formation or the aging due to non-uniform current density [7,10].

In some cases, the dispensing and wetting processes overlap. This often results from multi-stage dispensing processes in which the required target electrolyte volume is filled in successive partial volume increments. For instance, it occurs with hardcase cells in which the existing dead volume is smaller than the target electrolyte volume, or when there are limitations on the maximum filling quantity per dispensing stroke [16]. Since the dispensing times are generally shorter than the wetting times, the wetting process initiation is considered as the moment when the cell is sealed (cf. end of Figure 1b) [6].

Different parameters, such as the use of different pressure levels before and after cell closure, might influence this process step [12]. Thereby, time spent on these activities is grouped under the term "plant-related (auxiliary) time", which also includes transport to and from the plant. In contrast, the wetting process takes place outside the filling plant and is often further supported by temperature-controlled storage. In this context, the wetting degree serves as a parameter to indicate the percentage of pores that are wetted with electrolyte [17,18]—once reaching a sufficient level, formation initiates [19].

2.2. Classification in the Process Chain

The process chain of battery production decomposes typically into three stages: electrode production, cell production, and cell conditioning (cf. Figure 2) [20].

Electrode production involves the manufacture of coated electrode coils from the powdered starting materials along a series of production processes that significantly shape the wetting behavior of the electrodes. For example, the selection of the cell chemistry or material formulation with the different active materials, binders, and other additives at the molecular level determines how well the electrolyte interacts with these at the interfaces through the surface-free energies of these materials [21]. In the process steps of mixing, coating, drying, and calendering, the pore morphology of the electrode is set by various process parameters, which can be expressed, for example, in material parameters such as pore size distribution, pore volume, pore surface area, or tortuosity [22]. The pore morphology describes the essential influencing variable for determining the filling and wetting behavior related to the material [23,24].
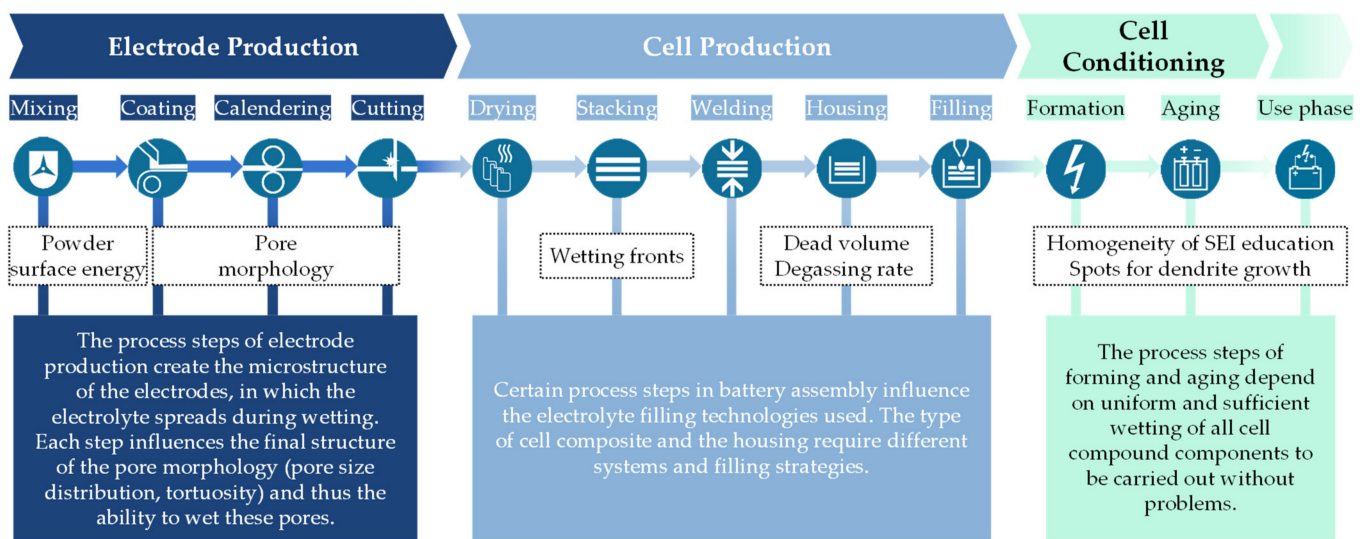
Figure 2. Schematic illustration of the process chain of battery cell production with the state-of-the-art process steps. The dashed boxes show the properties relevant to wetting, which are influenced by this particular process step.

Cell production is characterized by a large variance in the process technologies to be used [4]. Depending on the selected process technology, other technologies are either mutually dependent or mutually exclusive. Each upstream process technology potentially has an influence on the dispensing or wetting of the cell stacks. However, the effects are partially still unexplored. For example, current research barely touches on the influence of cutting, post-drying, and contacting on dispensing and wetting.

In the cutting process, the electrode material, which comes in the form of a coil, is cut into the correct shape for downstream packaging, such as cut-to-size coil material or sheets. It is not yet known whether the type and quality of the cut edge affects wetting, since this area is the main path for electrolyte transport from the dead volume into the pore system. Future research is needed to determine the effect of the packaging technique on wetting, as the electrode and separator materials are combined to form a cell composite. While jelly rolls only have two wetting fronts (the electrolyte spreads from the roll head and base to the center), stacked composites have four wetting fronts because the arrester foils do not present an in-plane obstacle, allowing the electrolyte to penetrate along each edge [17]. Similarly, the type of enclosure chosen has a significant impact on dispensing. Hardcase cells have significantly lower dead volumes and therefore require multi-stage dispensing procedures with wetting pauses in the double-digit minute range [16]. Softcase cells, on the other hand, often only require one dispensing step and the deformation of the housing due to the differential pressure between the cell interior and ambient pressure [25].

There are also research gaps in the post-drying process step, where the cell composite material is dried using low-pressure and temperature processes to reach a predefined, lower water content level [26]. The literature only provides indications on the influence of residual moisture on cell performance through reaction with the electrolyte [27–29]. In the contacting step, the electrodes and arrester tabs connect together, so that the electrode layers switch in parallel to form a robust current arrester to the outside without being apparently influenced by wetting or dispensing.

The electrolyte filling process, as described above, completes the battery cell assembly and transitions to cell finishing. Once sufficient wetting has taken place (achieved through storage time), formation is initiated followed by aging (cell conditioning).

2.3. Explanation of the Common Methods for Measuring Electrolyte Filling

There are several methods for characterizing the wettability of battery cell materials and whole battery cells, which involve measuring different values. Many of these methods

involve measuring a time-varying value that is proportional to the wetting, such as surface imaging [25,30], the wetting balance test of mass [31], or resistance in electrochemical impedance spectroscopy (EIS) [21,32]. These methods allow for the determination of wetting rates (dx/dt) or the absolute degree of wetting in optical methods [15,17,30]. Most measurements at the cell level and many measurements at the material level reference that relation.

Other measured quantities are those that are invariant with time, such as the contact angle between a solid, liquid, and gaseous material [33]. These measurements are typically only conducted at the material level. We explain them in detail in the following:

In the Wetting Balance Test, the wetting of cell composite materials is determined by the weight of the absorbed liquid. The measurement can be performed at specific points in time or continuously. In the former case, the horizontal cell composite material is brought into contact with a drop containing a specific amount of liquid, and the unabsorbed amount of liquid is removed and weighed after a waiting time using a balance. The characteristic value, electrolyte uptake [%]. is calculated from the difference between the material weight before (W_0) and after (W_1) the electrolyte uptake (cf. Equation (1)). The wetted material can then be stored under controlled conditions (temperature, pressure, humidity) to determine the electrolyte retention [%] at specific times through the equilibrium weight (W_X) (cf. Equation (2)). This indicates the opposite effect to electrolyte uptake [34].

$$\text{Electrolyte uptake} = \frac{W_1 - W_0}{W_0} \cdot 100\% \tag{1}$$

$$\text{Electrolyte retention} = \frac{W_x - W_0}{W_1 - W_0} \cdot 100\% \tag{2}$$

The continuous measurement is carried out with the aid of a tensiometer, in which the vertical material sample is brought into contact with a liquid reservoir at the lower end of the sample. The continuously increasing liquid weight m due to capillary effects is measured [31]. Based on the Washburn equation (cf. Equation (3)), the penetration rate (K) [$g \cdot s^{-1/2}$] is calculated from the measured mass–time data.

$$m = K \cdot \sqrt{t} \tag{3}$$

The Capillary Rise and the Droplet Spread Tests measure the spread of the liquid front (height h or area A) using a camera. The changing brightness of the wetted material shows the local wetting. The sorptivity, also known as permability (k) [$mm \cdot s^{-1/2}$], is used as a parameter (cf. Equation (4)).

$$h = k \cdot \sqrt{t} \tag{4}$$

In these tests, the samples are oriented differently (cf. Figure 3). While in the droplet spread test, a drop is placed on a horizontally lying sample (similar to the wetting balance test with a scale), the capillary rise test places the sample vertically in a liquid reservoir, similar to the test setup of the tensiometer.

Another method for determining the wettability of cell stack materials is the Contact Angle Measurement. Here, it is most common to use sessile drops, where a drop with a predefined amount of liquid is placed on the sample surface [33]. The contact angle (θ) [$°$] is the angle between the tangent to the drop surface at the transition to the sample surface and the sample surface itself. The wider the droplet spreads, i.e., the smaller the contact angle, the better the wettability of the sample material, as it is energetically better for the liquid to form new bonds with the sample surface on a molecular level than with itself. The structure of the surface also affects the formation of the contact angle [35,36]. For cell stack materials, it is difficult for a droplet to form consistently over time due to the seepage of the electrolyte into the material, making it difficult to apply this measurement method [37]. Therefore, this approach is suitable to characterize polyolefin separators,

which are known to have poor wettability due to their polyolefin base and the interaction with hydrocarbon-based solvents [38].
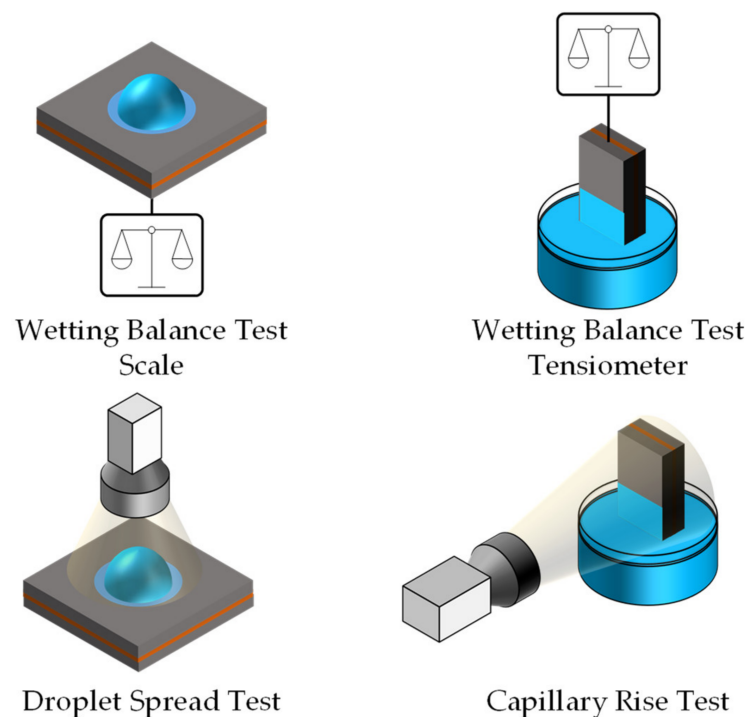

Figure 3. Schematic illustration of the common measurement methods used to measure the wetting properties of cell composite materials.

The other measurement procedures are predominantly used to investigate the wetting properties of battery cells, as shown in Figure 4. Another group of measurement techniques aims to visually map the wetting fronts in battery cells. Since the battery cells are surrounded by opaque enclosures during the dispensing process, radiative or emissive methods often support this purpose. One source of radiation deployed in this process is X-ray-based inspection, which serves to detect density differences. The intensity of transmissive radiation decreases as the transmissive material becomes denser. However, a contrast agent must be added to the electrolyte, otherwise the density differences between the electrodes and the electrolyte cannot be clearly distinguished [25,39]. The method visualizes in images the progress of the wetting fronts as well as areas where residual gas bubbles are located. Characteristic values such as the temporal change in the wetted area or the velocities of individual wetting fronts can be determined for all imaging methods. In contrast to X-rays, which interact with electron shells in atoms, Neutron Radiography excites the atomic nucleus of irradiated elements [30]. In this process, lighter elements such as hydrogen, lithium, or boron react more to this type of radiation than metallic elements, resulting in a change in detected radiation intensity for light elements. This allows for imaging of the wetting front of the hydrocarbon-based electrolyte.

Lock-In Thermography images the emitted thermal radiation generated by an oscillating current in the battery. Infrared cameras are used for this purpose [38]. The diffusion processes excited by the current flow can only occur in the wetted areas, which means that only these areas emit the thermal radiation. Thus, the time course of the wetting fronts can be displayed.

Figure 4. Schematic illustration of the common measurement methods for measuring the wetting properties of assembled battery cells.

Lock-in thermography images the emitted thermal radiation generated by an oscillating current in the battery. Infrared cameras are used for this purpose [34]. The diffusion processes excited by the current flow can only occur in the wetted areas, which means that only these areas emit the thermal radiation. Thus, the time course of the wetting fronts can be displayed.
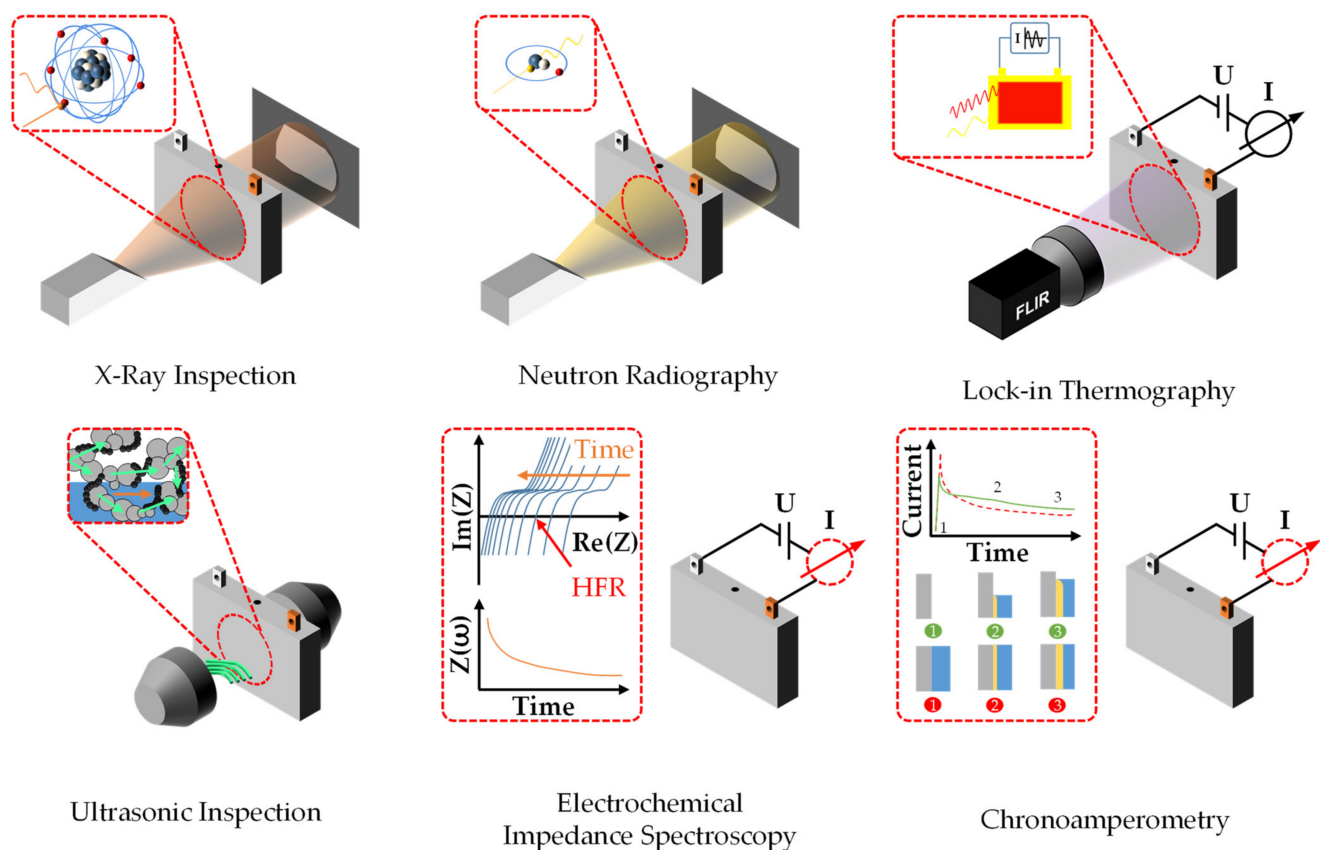
Furthermore, Ultrasound detects changes in characteristic parameters such as amplitude or sound velocity when it penetrates the battery cell stack, depending on the wetting state [40,41]. When traveling through the cell stack, ultrasound is transmitted through the cell via different paths (solid-state sound between the electrode and separator particles, airborne sound in gas, and liquid sound in electrolyte). Thus, the signal changes as soon as pores are filled with electrolyte instead of gas. This allows for determination of the wetting state over time, depending on the local detector's resolution.

Another common method for determining the wetting state of battery cells is the use of Electrochemical Impedance Spectroscopy (EIS). This method is particularly common for characterizing conditioned battery cells. It involves applying an AC current or AC voltage signal to the battery cell and calculating the impedance in each case from the inverse system response for a varying frequency. Since the resistance of the cell is inversely proportional to the wetted area, the change in several measurements can be used to make a statement about the wetting of the battery [21]. Currently, most EIS measurement and evaluation settings primarily map the macroscopic wetting of the separator and large pores [32].

The Chronoamperiometry method involves applying a voltage signal. It is based on differences in the measured current signal as a function of the solid electrolyte interface (SEI) formed. For this method, a voltage is applied to a cell with a fixed wetting time so that the negative electrode potential is in the range of 200 mV to 1.2 V, causing SEI formation without intercalation of Li-ions into the negative electrode. The measured time-dependent current signal (chronoamperogram) correlates with the reaction rate between the SEI and

electrolyte, which decreases with SEI thickness [37]. Since SEI formation reactions can only occur at wetted sites, chronoamperograms started at different wetting times differ. From the difference in chronoamperograms between a fully wetted cell and a cell to which the SEI formation voltage was applied immediately after filling, the time required for complete wetting can be determined. Using the geometric dimensions of the cell, the permeability (k) [mm·s$^{-0.5}$] of the cell stack can be calculated. [42].

3. Methods

To investigate the current state of research on dispensing and wetting, we conducted a systematic literature review (SLR) following the process proposed by Brendel et al. and Page et al. [43,44]. We included peer-reviewed scientific journal articles and conference papers to ensure the rigor of our review while also covering recent progress. We searched a variety of high-quality scientific databases from various adjacent disciplines in order to identify a wide range of potentially relevant contributions. Among these, we selected Scopus, which contains over 80 million documents and is referred to as the "largest abstract and citation database for peer-reviewed literature" [45]. We conducted the SLR in June 2022 using the following search phrase in the title, abstract, and keywords of the above-mentioned databases:

TITLE-ABS-KEY ("Electrolyte wetting" OR "Electrolyte filling" OR "Electrolyte distribution") AND ("Lithium-Ion battery" OR "batteries" OR "influence" AND "wetting").

The search query returned a total of 544 hits. Figure 5 presents the search and selection process in a PRISMA flow diagram as proposed by Page et al. [44]. Our decision to categorically exclude simulation studies from our literature review was based on several reasons. Firstly, we aimed to concentrate on studies that extracted data from pre-existing experimental studies on material process parameters and provide a contribution to an area where there is a research gap regarding review articles. Secondly, we viewed simulation as a distinct field that requires a foundation of overview regarding validation, which we intended to create. Lastly, we sought to avoid overburdening the complexity of our review by not mixing both experimental and simulation strands, given that the number of simulation studies in this field far surpasses that of experimental contributions to material process parameters and may overshadow them. After removing all 170 duplicates, 374 results remained for detailed analysis that we then systematically narrowed down to 39 finally labeled papers, as illustrated in the PRISMA diagram in Figure 5.

The middle column lists the number of reviewed publications at each step, the right column lists the documents added through backward search as well as the inclusion criteria for each selection step, and the left column shows the sequential removal of excluded publications after each process step based on our pre-defined exclusion criteria. While we only included articles written in English that dealt with the dispensing and wetting of electrolytes, the exclusion criteria contained duplicates, articles written in languages other than English for the full text, articles that purely treated process models without addressing influencing factors, and contributions that lacked a deeper consideration of the filling process to ensure alignment with our research aims. We applied a peer-reviewed screening process to strengthen the objectivity of the SLR. Following the suggestion of Bandara et al., we labeled the final selection of 39 articles using the software MAXQDA [46].

Our labeling system first divided the results into three categories: dispensing and wetting with a focus on material analysis (1), process investigation (2), or research on variations of preceding processes (3). We then exploratively discovered more detailed differences in the studies' findings, which we present in the results section. We created a joint and explorative derivation of the (sub-)labels following Mayring, which ensured a shared understanding for assigning labels during the further labeling process and analysis of the results [47].
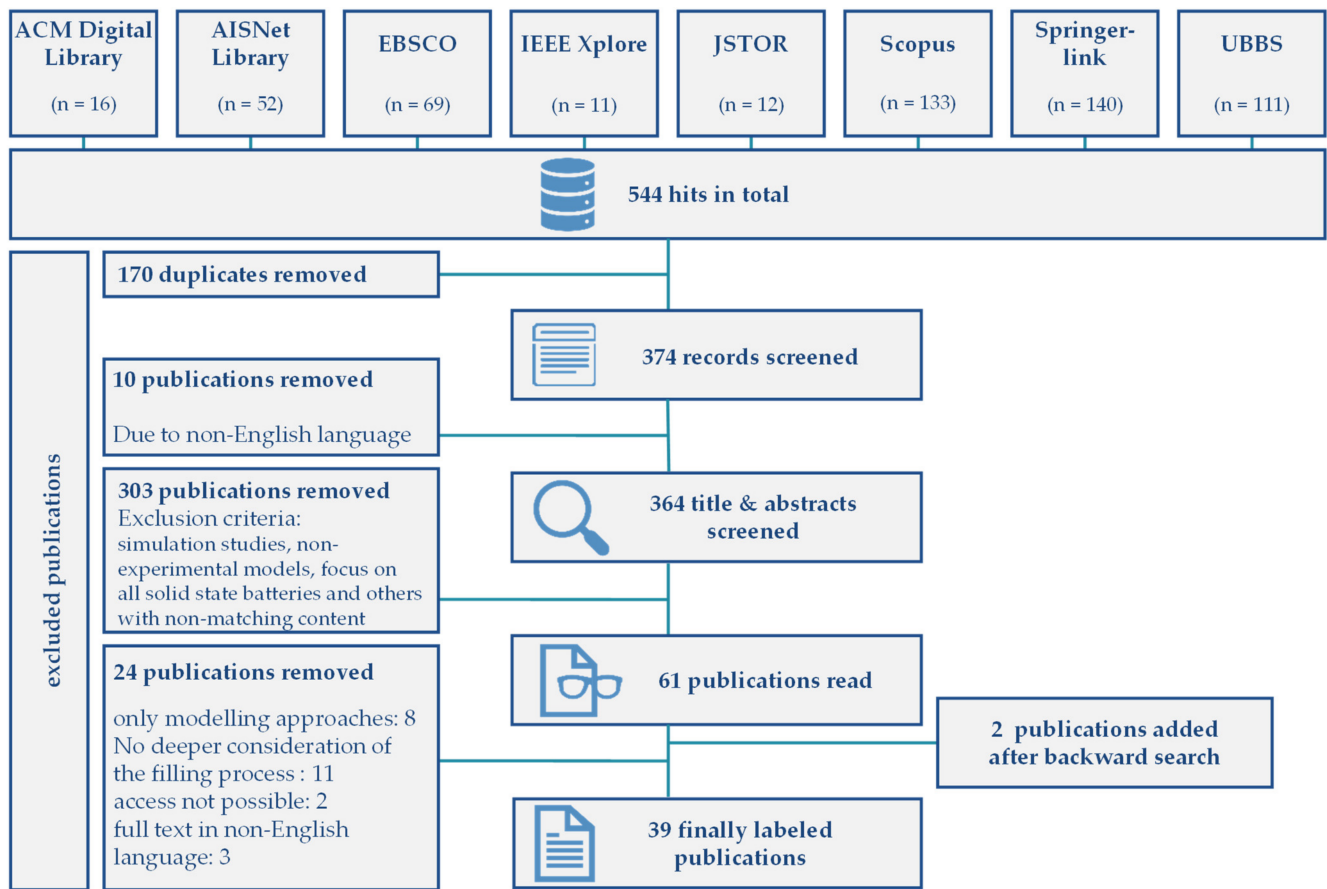
Figure 5. Paper selection process along the PRISMA statement.

4. Results

This section presents the obtained results. We organized the labeled contents of the publications into three categories and explained the main research topics within each category.

Figure 6 shows an increase in publications over the last ten years, with a peak in 2020, reflecting the growing importance of battery production over the last decade. This is particularly due to increasing efforts and political measures towards sustainability, which have a strong impact on the automotive industry. It is also notable that there has been an increasing number of contributions focusing on improved process parameters, as their efficient and effective design is crucial for the implementation of large-scale industrial production.

From 2011 to 2017, the focus was on material-centered publications related to electrolyte filling, such as studies on the wetting properties of newly developed materials and the effects of upstream process steps in electrode production, such as calendering or laser patterning, on the wetting properties of electrodes [48–50]. Since 2017 there has been a noticeable increase in publications investigating the electrolyte filling process itself, in addition to the development of new materials and the investigation of their wetting properties. Production-related issues are now also becoming a focus of scientific investigation.
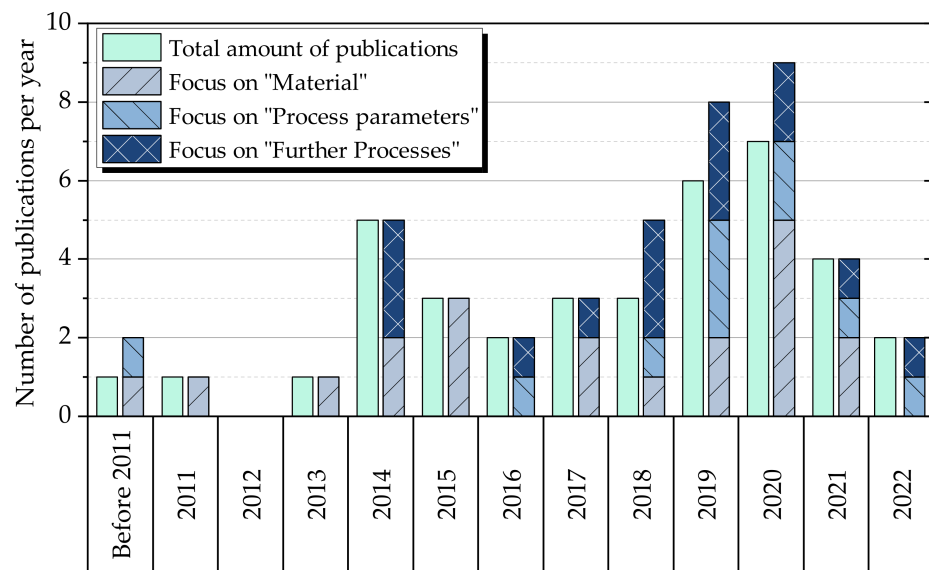
Figure 6. Illustration of the examined publications sorted by years. In addition, we assigned the publications to the corresponding main group labels. Since one publication can be assigned to various foci groups (e.g., material and further processes), its cumulation (right bar) sometimes exceeds the total amount of papers (left bar). The publications from 2022 include only those published up to the month of June.

We mapped the distribution of the labeled contents in a diagram (cf. Figure 7) according to their frequency. From the inside out, the sub-labels became more and more specific. Several labels of a contribution were therefore represented in several different labelings, so the sum of all labels exceeded the total number of examined publications by far. Approximately half of the publications (24/52) were related to materials, while roughly 20–25% were related to process parameters (13/52) and other processes (15/52).
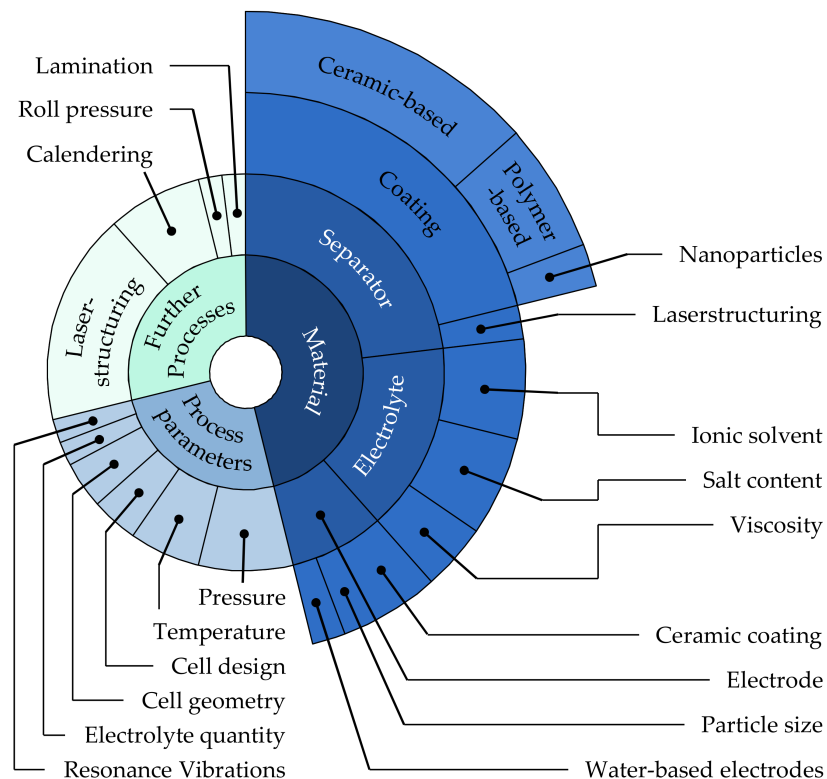

Figure 7. Representation of the labels and sub labels used in relation to their frequency of occurrence.

The category of Materials mainly referred to contributions that investigate the wetting behavior of different cell composite materials. The wetting behavior often characterized only one of many parameters used to dimension the cell composite materials (electrode, separator, and electrolyte). In most cases, however, the focus was on modifying the cell composite materials. The Process Parameters topic block included articles that specifically examined the electrolyte-filling process. Here, process parameters and certain product parameters such as cell design (hardcase vs. pouch cells) or cell geometry played a dominant role. The topic block of Further Processes dealt with research contributions on the influence of processes other than electrolyte filling on wetting. In addition to the divisions into topic blocks with corresponding sub-labels, we also indicated which level the research addressed: the Material Level, which included analyses of individual materials of the cell stack without a cell composite present; or the Cell Level, where analyses were performed on assembled cell composites. Tables 1–3 below illustrate the categorization made of the research articles selected and analyzed in full text.

Table 1. Overview of the publications assigned to the category material with a classification of the contents [21,31,34,37,38,42,50–63].

Studies	Parameter Electrode			Parameter Separator				Parameter Electrolyte			Measurement Method										Level	
	Ceramic Coating	Water-based Electrodes	Pore Size Structuring	Ceramic-based Coating	Polymer-based Coating	Laser-structured	Nanoparticles	Ionic Solvent	Salt Content	Viscosity	Capillary Rise	Wetting Balance	Droplet Spread	Contact Angle	Laser-based	X-Ray-based	Neutron Radiography	Chronoamperometry	Thermography	EIS *	Material Level	Cell Level
An et al. 2014 [50]				×	×						×	×									×	
Beyer et al. 2020 [37]				×							×			×		×					×	
Bolimoswka et al. 2017 [51]								×							×				×		×	
Davoodabadi et al. 2018 [52]		×												×							×	
Davoodabadi et al. 2019 a [53]						×	×						×								×	
Davoodabadi et al. 2019 b [54]		×											×								×	
Davoodabadi et al. 2020 [55]				×									×								×	
Fang et al. 2011 [56]							×						×								×	
Jang et al. 2021 [57]			×										×						×		×	
Kühnel et al. 2013 [31]									×	×		×							×		×	×
Man et al. 2014 [58]				×									×								×	
Peter et al. 2020 [42]	×			×										×			×					×
Pröll et al. 2015 [59]						×							×								×	
Schilling et al. 2020 [38]				×								×	×					×			×	×
Sun et al. 2021 [60]								×					×								×	
Wang et al. 2015 [61]					×							×	×								×	
Wang et al. 2017 [62]				×							×	×	×								×	
Wang et al. 2020 [31]				×				×	×			×	×								×	
Wu et al. 2004 [21]													×								×	
Yoo et al. 2015 [63]					×						×		×								×	

*Abbreviation: EIS = Electrochemical Impedance Spectroscopy

Table 2. Overview of the publications assigned to the Process Parameters category with a classification of the contents [11,15–17,21,25,30,42,54,64].

Studies	Parameter Process — Pressure	Electrolyte Quantity	Temperature	Cell Geometry	Cell Design	Resonance Vibrations	Measurement Method — Capillary Rise	Wetting Balance	Droplet Spread	Contact Angle	Laser-based	X-Ray-based	Neutron Radiography	Chronoamperometry	Thermography	EIS *	Level — Material Level	Cell Level
Davoodabadi et al. 2019 a [54]			✕						✕								✕	
Günter et al. 2019 [11]		✕													✕			✕
Günter et al. 2020 [17]				✕									✕					✕
Günter et al. 2022 [16]	✕	✕											✕		✕			✕
Jin et al. 2021 [64]						✕	✕				✕	✕					✕	
Knoche et al. 2016 [30]	✕												✕					✕
Peter et al. 2020 [42]			✕	✕						✕				✕				✕
Schilling et al. 2019 [25]			✕									✕						✕
Weydanz et al. 2018 [15]	✕			✕			✕						✕					✕
Wu et al. 2004 [21]	✕															✕		✕
*Abbreviation: EIS = Electrochemical Impedance Spectroscopy																		

Our analysis showed that a significant proportion of the publications focus on the wetting properties of materials at the material level, particularly the modification of poorly wettable polyolefin separators. These modifications in the majority of coatings made of ceramics or other polymers are intended to improve the properties of separators in general, such as thermal or mechanical durability. These changes to the surface often also result in an improvement in wetting ability, as the polar and disperse components of the free surface energies of these coatings better match those of the electrolyte solvents.

Research also focused on the modification of electrolyte composition, primarily involving carbonate-based electrolytes with LiPF6 as the conducting salt [31,34,54]. Only a few studies, such as the work of Davoodabadi et al., investigated specific variations in electrode formulation, using a water-based binder in their formulation [52,54]. Simple material characterization methods, such as the droplet spread test or contact angle measurement, were commonly used. In contrast, there was a lack of investigation into the transfer of findings to the cell level through cell performance tests, indicating a research gap.

Our analysis also revealed several publications that addressed the main process parameters of temperature and pressure [15,16,54]. While temperature experiments were conducted at both the material and cell levels, they were never conducted in the same experiment, leaving a lack of information on the possible mutual influence of the process parameters on each other and the influence of different material parameters. Again, the material and cell levels were only investigated in isolation without analyzing the transfer of findings from the material level to the cell level.

Table 3. Overview of publications assigned to the category Further Processes with a classification of the contents [7,15,42,49,52,54,65–72].

Studies	Parameter Further Processes				Measurement Method										Level	
	Calendering	Laser-structuring	Lamination	Rolling Press	Capillary Rise	Wetting Balance	Droplet Spread	Contact Angle	Laser-based	X-Ray-based	Neutron Radiography	Chronoamperometry	Thermography	EIS *	Material Level	Cell Level
Alaboina et al. 2017 [65]	✕							✕							✕	
Berhe und Lee 2021 [66]		✕						✕							✕	
Davoodabadi et al. 2018 [52]	✕							✕							✕	
Davoodabadi et al. 2019 a [54]	✕							✕							✕	
Habedank et al. 2019 [67]		✕									✕					✕
Kaden et al. 2020 [68]			✕		✕	✕							✕		✕	✕
Kleefoot et al. 2022 [69]		✕			✕		✕								✕	
Peter et al. 2020 [42]		✕										✕				✕
Pfleging & Pröll 2014 [7]		✕			✕										✕	
Pfleging et al. 2014 [49]		✕			✕										✕	
Pfleging et al. 2016 [70]		✕								✕					✕	
Pfleging 2018 [71]		✕													✕	✕
Schilling et al. 2018 [72]				✕												✕
Sheng et al. 2014 [48]	✕							✕							✕	
Weydanz et al. 2018 [15]		✕									✕					✕
*Abbreviation: EIS = Electrochemical Impedance Spectroscopy																

The most commonly used measurement methods were electrochemical impedance spectroscopy and neutron radiography. Cell design and geometry also played a role in the dispensing and wetting conditions between pouch and hardcase cells. However, the evacuation process and the pressure it set were not fully considered in the literature. It is unclear whether the pressure applied on the process side also affects the pore system or just the dead volume of the cell. This is especially important for hardcase cells, which have narrow, coiled designs, where the diffusion paths for gas from the pores correspond to half the cell composite height.

The cell geometries discussed in the literature showed little variation. While the size and number of compartments may vary, the cell geometries often have similar aspect ratios with the same length-to-width ratio. With the trend in the automotive industry is towards blade cells for use in the cell-to-pack battery, the impact of varying the electrode area and shape, particularly the aspect ratio, has not been explored.

The analyzed literature covered various studies on upstream and downstream processes. Some contributions focused on the influence of calendering and laser structuring processes on the wetting properties of electrodes. Calendering, as the final process step

in electrode manufacturing, significantly affects the pore morphology of the electrode. However, there is a lack of a comprehensive view that takes into account the influence of mixing, coating, and drying parameters on the resulting pore morphology in addition to calendering parameters. This would allow for more general conclusions to be drawn about the relationship between the electrode manufacturing process parameters, pore morphology, and wetting behavior of the electrode. Additionally, it would be useful to consider the materials used, as they have a major influence on both the resulting pore morphology and wetting properties.

Laser structuring is an additional or alternative process step in the manufacturing process chain that is used to improve ion diffusion into electrodes, such as for fast charging processes. However, this process step also leads to a decrease in specific volumetric capacity [67]. The introduced structures also provide additional channels for electrolyte to spread better in the direction of the electrode thickness and create punctual starting points for wetting in otherwise remote regions of the electrode. Many publications studied the influence of this process step on wetting at both the material and cell levels. Other articles occasionally addressed other processes, such as lamination of electrode-separator composites, or process improvements for electrolyte filling processes such as roll pressing, in which the electrolyte is distributed in the filled and sealed cell by a roller [68,72].

The bar chart in Figure 8 illustrates the frequency with which different measurement methods were used in the publications studied. It distinguishes between investigations at the cell level (in dark blue) and those at the material level (in light blue). While contact angle measurement and wetting balance tests are commonly used at the material level, electrochemical impedance spectroscopy and neutron radiography are most frequently used at the cell level. The choice of measurement method is often dictated by the level at which it operates; contact angle measurement can only be performed between a solid and a liquid, so it cannot be used to image the cell level. Similarly, chronoamperometry and electrochemical impedance spectroscopy require a complete cell, which precludes measurements on individual cell stack materials. Therefore, no dominant measurement method emerged.
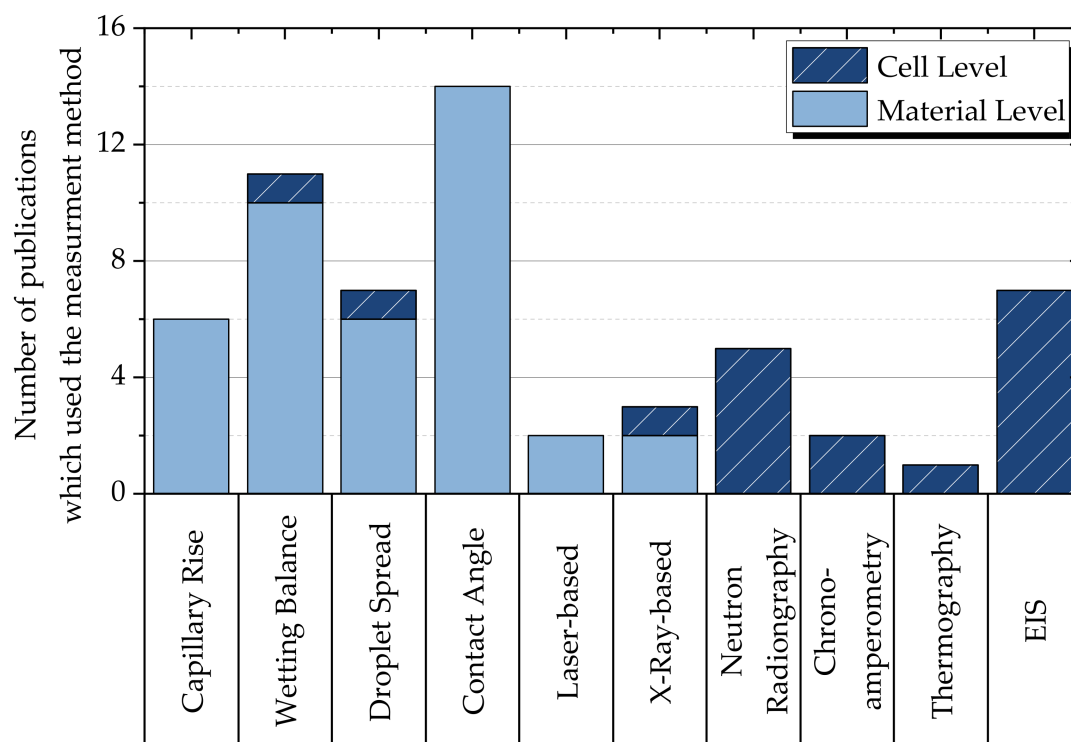

Figure 8. Illustration of the number of measurement methods used in the publications examined, with a classification of the measurements into material and cell levels.

However, we identified certain recurring patterns: Wetting measurements were generally limited to either the material or cell level. There was a strong tendency to use these methods at the material level when investigating modifications of cell stack material (e.g., coatings on separators, modification of electrode material, post-mortem analyses). These methods have the advantage of requiring a small amount of material and a simple measurement setup, which makes them cost-effective. However, they do not take into account reciprocal influences between materials. Measurements at the cell level, on the other hand, were used when investigating process parameters and the reciprocal material influences within the entire cell compound. These investigations provided a more realistic depiction of the "electrolyte filling" process through in operando measurements. A gap in the research is the lack of experimental investigations that address both levels at the same time and allow for transfer from the material level to the cell level. If such a transfer were successful, it would have great potential to draw conclusions from material level research for effects at the cell level, enabling faster, more cost-effective, and more precise predictions.

5. Discussion

In the previous section, we provided a detailed overview of the material and process-specific influencing parameters investigated in the literature and classified them by the material or cell level and the measurement methods used. As previously mentioned, current research lacks the transfer of results from the material level to wetting behavior at the cell level. However, initial measurements of wetting properties at the material level are being conducted for new material developments or process changes in electrode production. These data must be used effectively. One limiting factor is resources (material and analytical capacities) available, which makes it difficult to conduct test series with multi-layer, large-format battery cells, as they require a larger sample set of material. In addition, setups for measurements on materials are generally easier and cheaper to obtain. However, knowledge gained from measurements at the material level cannot be directly transferred to the cell level, as reciprocal effects between the materials are not taken into account. For example, if the separator is a bottleneck in the wetting at the cell level, improving the wettability of the electrodes may not necessarily result in proportionate improvement in the wetting of the cell. This highlights the need to transfer knowledge gained at the material level.

The results of this study have identified several research gaps that should be given special attention in the future. First, the literature review revealed that the measurement methods used at both the material and cell levels are very heterogeneous. There is a lack of research comparing these different approaches or proposing a unified measurement procedure combining best practices and advantages from each approach. While studies at the material level tend to focus on the characterization of cellular materials or their modification, with wetting being just one of many sub-aspects, investigations at the cell level primarily focus on the wetting process itself. Due to the significant differences in measurement principles and variables used at both levels, it is often difficult to compare the results of these studies. In particular, many measurement methods cannot be applied at both levels, so any comparison between them must be made using a derived parameter such as the sorptivity or permeability (k) $[mm \cdot s^{-1/2}]$, which indicates the dispersion velocity of a liquid in a porous medium.

At the material level, wetting properties are often determined using the wetting balance test, capillary rise test, droplet spread test, or contact angle measurement. However, most measurements at the material level applying these methods are conducted at ambient pressure and room temperature. While it is possible to vary the temperature to some extent with these methods, it is more difficult to investigate variations in ambient pressure during wetting. Radiographic methods and ultrasound could be used to study wetting at the material level, but since the material samples are not enclosed, these methods are either unnecessary or too costly compared to optical inspection using a camera. Chronoamperiometry and electrochemical impedance spectroscopy, on the other hand, rely on the

evaluation of a current signal resulting from a given voltage as the system response of a battery cell, so they can only be performed at the cell level. Similarly, lock-in thermography is limited to being performed on a functioning battery cell.

In summary, the accuracy or resolution required and the available resources (e.g., time, personnel, equipment) should be considered when selecting measurement methods for wetting properties. If the specific wetting properties of interest require specialized equipment, it may be necessary to use a more specialized measurement method. On the other hand, for investigations of new materials or material modifications, a measurement method that is easy to use and that provides a high level of information about the measured variable may be more useful. We summarized an overview of the requirements and capabilities of each measurement method, as shown in Table 4.

Table 4. Overview of the measurement methods used to determine wetting properties.

Measurement method	Equipment	Level	Evaluation parameter	Measurement principle
Capillary Rise	Camera	Material	Height [mm] Sorptivity [$mm \cdot s^{-1/2}$]	Visualization of the wetting front against gravity
Wetting Balance - Uptake Method	Scale (& oven)	Material	Electrolyte uptake [%] Electrolyte retention [%]	Change in weight at specific points in time
Wetting Balance - Washburn Method	Tensiometer	Material	Mass [g] Penetration rate [$g \cdot s^{-1/2}$]	Continuous change of weight
Droplet Spread	Camera	Material	Wetted Area [mm^2] Sorptivity [$mm \cdot s^{-1/2}$]	Visualization of the wetting front in the plane
Static Contact Angle	Drop Shape Analyser (DSA)	Material	Contact angle [°]	Contact angle of the drop on surface
Chronoamperometry	Potentiostat	Cell	Current difference [mA]	Measurement of the current change during SEI formation
Thermography	IR Camera, Potentiostat	Cell	Wetted Area [mm^2] Sorptivity [$mm \cdot s^{-1/2}$]	Visualization of the wetting front by emitted thermal radiation
EIS	Potentiostat	Cell	High-frequency resistance [$m\Omega$] Impedance at 1 Hz [$m\Omega$]	Measurement of impedance change
Ultrasonic	Ultrasonic transmitter & receiver	Material & Cell	Transmitted Amplitude [dB] Sound Velocity [$mm \cdot s^{-1}$]	Measurement of amplitude and frequency change
X-Ray-based	X-ray source (plant)	Material & Cell	Wetted Area [mm^2] Sorptivity [$mm \cdot s^{-1/2}$]	Visualization of the wetting front by excitation of the dissolved contrast agent
NR	Neutron source (reactor)	Material & Cell	Wetted Area [mm^2] Sorptivity [$mm \cdot s^{-1/2}$]	Visualization of the wetting front by excitation of the hydrogen

In this paper, the wetting balance test (operated by a scale) and the droplet spread test (operated by a camera) are considered because these methods are similar in their design and involve the injection of droplets onto a horizontally mounted sample (cf. p.6). A variant of the droplet spread test, described by Davoodabadi et al. and shown in Figure 9, involves depositing the material on a transparent polyethylene terephthalate plate and sealing it with an adhesive tape made of polyethylene [53]. The adhesive tape has a circular recess as a starting point for wetting, which improves the circular propagation profile and allows for the evaluation of the wetting parameter using machine vision while also eliminating evaporation effects. If the sample is an electrode coated on a non-transparent arrestor or on both sides, the camera test can also be performed from the top side.
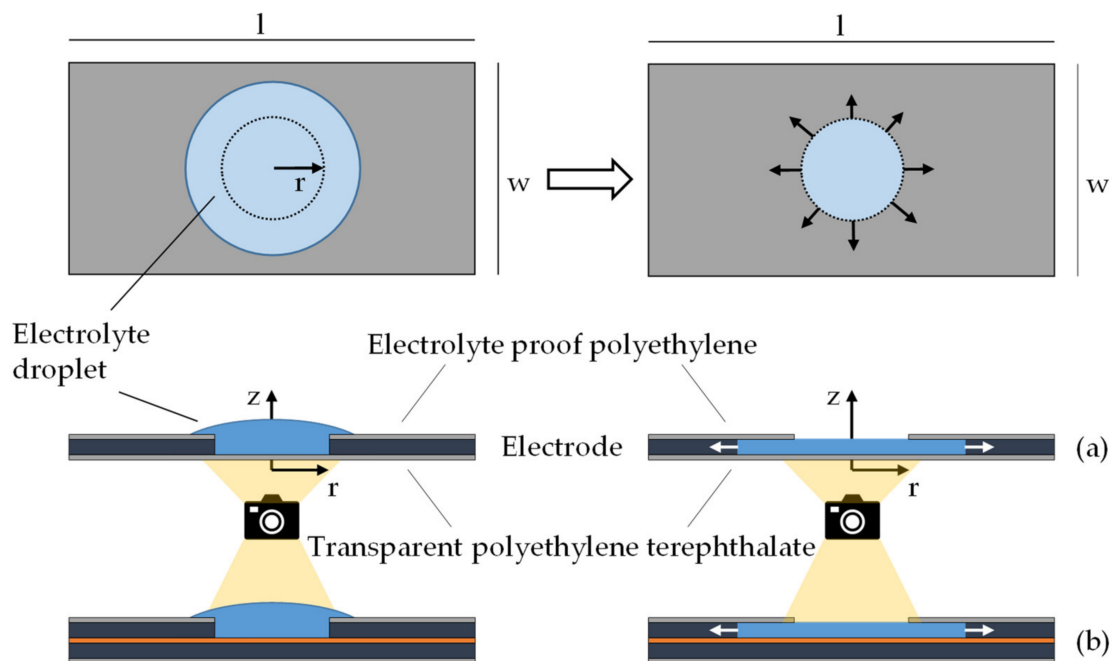
Figure 9. (**a**) Illustration of the measurement method used in the publication by Davoodabadi et al.; (**b**) an extension of the method for double-sided coated electrodes. [53].

The wetting balance test using a tensiometer or the contact angle measurement have relatively high initial costs for the necessary equipment, making them less suitable for wetting measurements as a secondary investigation.

For the analysis of factors that influence the dispensing or wetting of entire battery cells, specialized measurement setups are often necessary due to the high acquisition costs for the equipment and the risk of hydrofluoric acid formation if the electrolyte comes into contact with the humid ambient atmosphere in which the equipment is often located. In addition, it is important to have good detectability of the macroscopic wetting processes in this application. Radiographic methods, such as neutron radiography, offer particularly good possibilities for this, but neutron radiography is only available at a few locations due to the limited availability of the radiation source. X-ray-based methods have greater facility availability, but they also have proven to be expensive in terms of facility acquisition and require modification of the electrolyte. The effect of adding contrast agents on wetting and performance characteristics has not been investigated.

Electrochemical impedance spectroscopy, a well-known and widely used technique in cell diagnostics of formed cells, can be used as a measurement technique for investigating the dispensing and wetting processes at the cell level. This is because the potentiostats used for cell diagnostics, which are capable of performing electrochemical impedance spectroscopy, are often available as infrastructure to benefit from. However, this measurement method only maps the separator wetting or the macroscopic wetting of the cell composite by measuring and evaluating the high-frequency resistance. There are no methods available for determining the microscopic wetting of the pore system, which may involve a measurement and corresponding evaluation of low frequencies.

Moreover, all these mentioned measurement methods are designed or tested for use in experimental setups on a laboratory scale. In particular, transferring these methods to a measurement concept at the cell level for series production, which is a current focus of scale-up, is an area predestined for future research, since the transfer of wetting properties from the material to the battery cell level involves reciprocal material and process influences that cannot be depicted at the material level alone. One challenge we see in this context is the selection of the type of measurement principle (e.g., relative change from the initial measurement point). These principles are often based on measurements of a changing

quantity, such as in electrochemical impedance spectroscopy, where the wetting state (e.g., degree of wetting) is inferred by the fact that the measured quantity has changed to a corresponding value compared to the starting value. In series production, a measurement method used to inspect the degree of wetting and classify it as good or scrap should have a measurement duration that is as short as possible in order to minimize the time the measurement equipment is occupied and maximize throughput. Measuring an absolute quantity, which is independent of the starting quantity, may be more useful in this context, such as the proportion of the wetted electrode stack for radiographic methods.

For relative measurements, calibrating the produced battery cells may be a suitable solution. However, valid calibration in this context requires very stable production, as even small deviations can result in changes and invalidate the calibration value. While research may provide useful approaches in this area, practical support is needed to achieve the necessary production capacity and data quantity and quality to derive reliable values.

An ultrasonic method could be an interesting option for mass production. It is crucial in the course of this to properly transfer and remove ultrasound from the battery cell, specifically the way in which the cells are positioned between the measuring heads. This procedure might be particularly appealing for use in mass production if the battery cell remains in the ambient atmosphere instead of in a liquid medium, as is the case with currently available applications. Our analysis also briefly examined the effects of individual production steps, such as calendering, on the wetting properties of electrodes during their production. However, we did not conduct a systematic investigation into the overall impact of the entire electrode production process on the wetting properties. It is necessary to thoroughly understand the mutual effects of the individual steps on the final pore morphology and their influence on wetting. There are also processes that are commonly used in the industry but that have received little or no research attention. There is great potential for systematically studying and improving these processes with regard to their effects on the electrolyte filling process. For example, we could research the roll pressing process, which has not been extensively studied, in conjunction with the application of heat to improve the wetting behavior of the electrolyte and increase the speed and efficiency of the roll pressing process.

With our literature review, we identified research gaps and presented the current state of research in the field. However, we would like to admit limitations of our study, which we will explain in more detail below. First, we queried the databases in June 2022, so that more recent publications are not part of our analysis. For example, an article published in October 2022 demonstrates an improvement in wetting behavior due to plasma activation of the electrodes. In this process, the surface of the electrode material is treated with plasma before the packaging process in order to accelerate the absorption of the electrolyte and improve the C-rate capability in a subsequent application [73].

Second, our search terms and screening process resulted in the exclusion of publications on modeling and simulations of wetting processes, as these were not the focus of our investigation. While models and simulations can be a useful way to represent dispensing and wetting at different scales, validated models might depict a cost- and material-efficient way to investigate different materials and their combinations [74,75]. Future studies could particularly focus on simulation studies, building upon our review to then cover the following key research questions:

- What types of models and simulations are applied, and what computational algorithms are employed?
- At what level or scale are dispensing and wetting represented in these models?
- Are there simulations that focus on the wetting processes in the pore systems of cell composite materials, the dispensing process in the housing, or the cause-and-effect relationships in the overall process?
- What are the inputs and outputs of the respective model?
- What resources or computing power are required for a simulation?
- In what framework have these models been validated?

One possible use case for these publications is the development of a process model for transferring material-specific wetting from the material level to cell wetting, or for comparing existing approaches. Cell level wetting has many complex structure–process–property relationships beyond the characteristic values obtained at the material level. By means of a process model, this complexity can be mapped in a systematic way by capturing property relationships, providing a starting point for further investigations of the influences of these properties.

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