



Review

Comprehensive Assessment of Novel Technologies for Electrolyte Filling in Lithium-Ion Battery Production

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Abstract: Electric vehicles play a pivotal role in the decarbonization of the mobility sector. However, their success depends on low-cost, high-performance batteries, requiring continuous optimization of their production processes. Electrolyte filling is a critical and costly bottleneck in the cell assembly and influences the quality and safety of the cells, offering great potential for identifying process optimizations. The aim of this study is to complement existing studies by analyzing and evaluating novel technologies for electrolyte filling and thus to provide guidance for industry and science. A systematic literature and patent search led to the identification of sixteen relevant technologies. These were evaluated by a group of experts from the scientific community to identify the most promising technologies. As a result of this evaluation, five technologies emerged that were assessed as positive compared to the state of the art. Overall, the results of this study indicate that the dominating trend in electrolyte filling will be direct pressurization of the battery cells with increasing pressures. Apart from this trend, no other fundamentally new process technologies for industrial use are currently foreseeable. Our findings indicate that both academics and practitioners should focus future research and industrial efforts on optimizing and understanding the current process.

Keywords: electrolyte filling; lithium-ion battery; battery production; filling; degassing



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1. Introduction

In the wake of advancing global warming, the decarbonization of our society is becoming increasingly urgent. The transportation sector accounted for about 20% of global CO₂ emissions in 2021, with passenger cars accounting for 39% of those emissions [1,2]. In addition to shifting to less carbon-intensive modes of transportation, replacing internal combustion engine vehicles (ICEVs) with electric vehicles (EVs) lowers emissions [3]. The success of EVs relies on low-cost batteries with high performance. Reducing production costs is also crucial for the profitability of cell manufacturers. In turn, access to affordable battery cells is a decisive location factor for the value creation of electric vehicles at the respective business location [4]. To produce the battery cells at ever lower cost, the production processes must be continuously optimized. The production of battery cells can be divided into three main processes: electrode production, cell assembly and cell finishing. In electrode production, anodes and cathodes are produced. First, the respective materials are mixed and coated onto current collectors. The electrodes are then dried and compressed using a calender. Before being transferred into the cell assembly, the electrodes are slit to the appropriate length and width and are vacuum-dried. In cell assembly, the electrodes are either separated and stacked or wound together with the separator to form a cell body.

The electrodes are contacted, and the cell body is placed in a cell housing. After packaging the electrolyte filling takes place. Cell closure marks the end of cell assembly. Cell finishing starts with soaking and forming, in which the cells are charged and discharged for the first and repeated times. The cells are then aged under controlled conditions before they are subjected to a final quality control and sorted accordingly [5].

From these production processes, electrolyte filling accounts for around 5.5% of production costs, of which around 65% is attributable to materials and 35% to depreciation, labor, energy, capital and other costs [5]. With a throughput of 0.6 to 0.7 GWh/a per electrolyte filling system, the process is a bottleneck in cell assembly and must be highly parallelized to keep up with the throughput of a gigafactory. In addition, electrolyte filling is critical for the quality and safety of the cell, as complete wetting of the cell must be ensured to maximize performance and prevent formation of high surface area Li deposits due to local current differences [6]. These metallic microstructures formed during charge on the anode can lead to short circuits [7]. In summary, there is great potential for identifying process optimizations in the electrolyte filling process.

Electrolyte filling subsists two sub-processes: Filling and Wetting. In the filling sub-process, the electrolyte is dosed into the dead volume of the battery cell. The dead volume refers to the volume within the cell housing that is neither occupied by the cell stack nor by other cell components. In the wetting sub-process, the electrolyte penetrates the pores of the porous electrodes and separator. As the overall process is called electrolyte filling and the sub-process is called filling, it can be difficult to differentiate between the two terms. In literature and patents, other terms such as dosing, dispensing, or injection are sometimes used to make the difference between the sub-process and overall process clear [8–10]. Nevertheless, the meaning usually only becomes clear through the context.

Existing literature mainly deals with wetting properties affecting both sub-processes of electrolyte filling. In a recent publication, Kaden et al. analyze articles and conference papers dealing with the experimental investigation of wetting properties. In doing so, they addressed studies on material properties, process parameters and further processes as well as the associated measurement methods. The study shows that in previous research there has been hardly any transfer of the results on wetting behavior at the material level to the cell level. In addition, the measurement methods of material and cell level differ from each other, so that results are not comparable [8].

Knoche, for example, dealt with the filling sub-process in his publications [9,11–13]. In the process model Knoche et al. they developed for the electrolyte filling process of lithium-ion batteries, focus lies on the dosing process by means of a morphological box [11]. They use the morphological box to describe specific elements of the electrolyte filling process in detail and derive their attributes. With their process model, they aim to support the conception and design of the electrolyte filling process and machine while also providing a base for further empirical research into the interdependencies and cause-and-effect relations in the process. In another publication, they visualize the electrolyte filling process using neutron radiography [12], discuss the observed process phenomena like gas entrapments and draw conclusions for cell production. Specifically, it is emphasized that the wetting time depends on the initial degree of wetting and exponentially on the initial rate of electrolyte absorption. Therefore, a well-designed filling sub-process can reduce the wetting time [12]. Lastly, in their publication from 2015 [9], they qualitatively describe the various influences of different dimensions such as cell stack, housing, machine or environment on the wetting degree during the electrolyte filling process. The course of pressure during and after dosing, the sealing process and the avoidance of humidity, for example, are identified as some of the most influential factors [9].

While they describe some individual technologies and a process model, it is rather generalized and dates back several years. Since the last publication on electrolyte filling, which Knoche contributed to in 2018 [14], new technologies have been developed. However, existing studies mainly focus on the scientific aspects of generating understanding of the processes on small cell formats in laboratory environment [15–23]. Studies with respect to industrial applications and focus on large cell formats are rare. Nevertheless, knowledge on scaling effects and process optimization for industrially applicable cell formats are inevitable for process optimization and deriving recommendations for future process development. Our aim is to complement existing studies by analyzing and evaluating novel technologies and thus to provide guidance for industry and science. To this end, a systematic literature and patent search was conducted, the results filtered, and then evaluated by a group of experts on the basis of various criteria. It turns out that industry has numerous technology ideas for optimizing the process in terms of speed, flexibility, safety, and quality. Among them we identify several as eminently promising to contribute to an optimization of the process.

This article focuses primarily on the filling sub-process and assesses novel technologies described in publications, patents, and expert interviews. To this end, we will first present the current state of the art of electrolyte filling in Section 2. We will then describe our methodological approach in Section 3, before describing and evaluating the novel technologies in Section 4. In Section 5, we summarize our results and assessments and point out potential future work.

2. Research Background

A variety of approaches may be employed in the implementation of electrolyte filling in battery cell production. This section offers an overview of the current state of the art. The process of electrolyte filling is first delineated. Subsequently, the factors that affect the duration of electrolyte filling are examined. Following this, the technical implementation of the filling sub-process is explained, with a particular focus on pressurization, before some concluding remarks are made.

Figure 1 shows two exemplary sequences of both sub-processes of electrolyte filling. The filling sub-process refers to the period where electrolyte is dosed into the dead volume of the battery cell. The dead volume comprises the free volume between the cell body and the cell housing. The cell body consists of numerous porous electrode and separator layers as well as impermeable metallic layers (current collector) and other inactive components. During the wetting sub-process, the electrolyte penetrates the porous layers. Since the penetration already begins when the electrolyte comes into contact with the cell body, the two sub-processes overlap as indicated by the mixed colored areas in Figure 1 which makes a separate consideration of the sub-processes difficult.

Sealing the cell completes the overall electrolyte filling process. One or multiple filling sub-processes can take place within this process window but only one wetting sub-process. Scenario 1 shows one filling sub-process accompanied by the wetting sub-process. Scenario 2 shows two filling sub-processes with a waiting time in between, during which only the wetting sub-process takes place. This waiting time is necessary for large hardcase cells, where the dead volume is smaller than the pore volume of the cell body and filling in one step is therefore impossible. In any case, the wetting sub-process will also continue after cell closure in the soaking process and only ends before the start of formation [8].

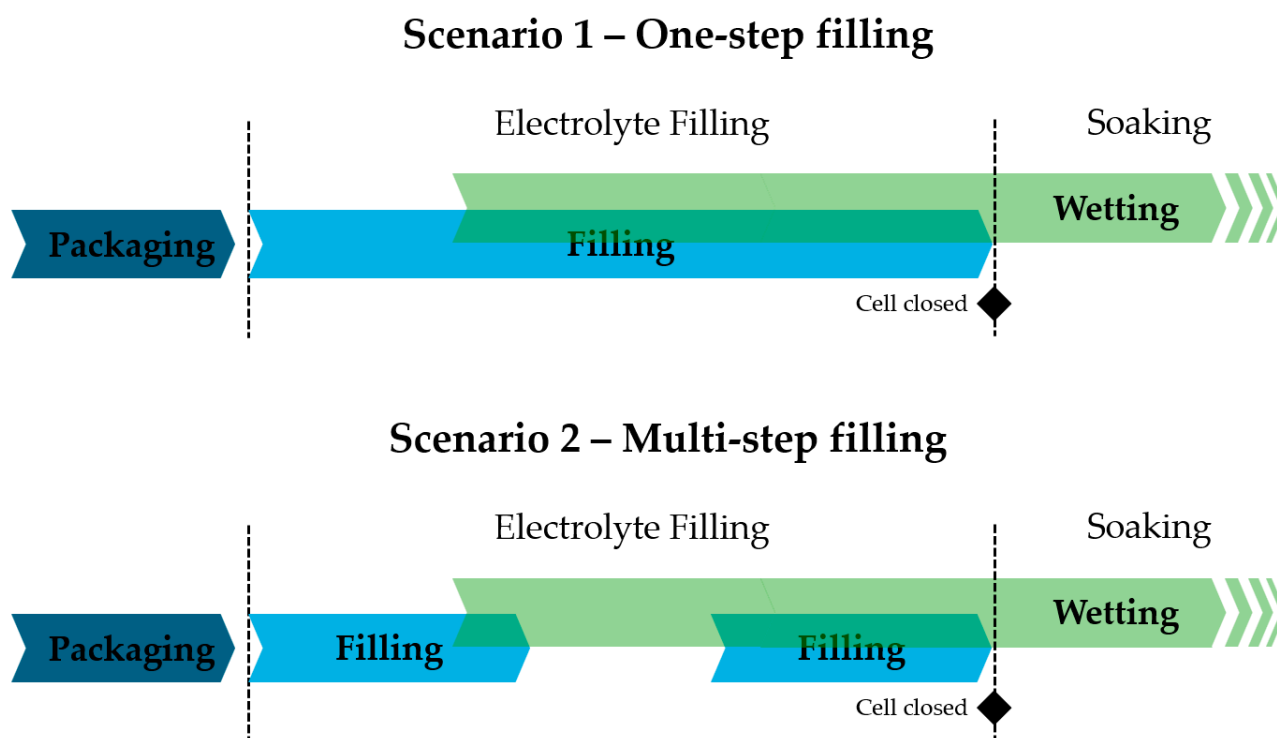


Figure 1. Diagram of the electrolyte filling process with its sub-processes. Scenario 1 with one filling iteration and scenario 2 with two filling iterations. Based on Kaden et al. [8].

The aim of the filling sub-process is to fill in a defined amount of electrolyte as quickly and precisely as possible, which is required to wet the porous cell stack completely and thus ensuring cell functionality and safety [8,9].

The duration of the filling sub-process is related to the cell design and can vary from a few seconds to almost an hour according to experts from industry and academia [6]. However, the availability of data and information is very limited, especially for industry-relevant scales. The wide range in process duration is due to the numerous influencing factors. The relationships between these factors and the duration of the filling sub-process are detailed in the following. Factors regarding the cell design, such as the electrolyte quantity, dead volume, type of housing, wettability of the materials, porosity and assembly type are discussed first. The paragraph will conclude with a discussion of factors regarding process parameters such as temperature and pressure. First of all, the required electrolyte quantity and the rate at which the electrolyte is pumped into the cell are decisive. The electrolyte quantity primarily results from the pore volume and therefore from factors such as cell capacity and porosity. Moreover, a surplus of electrolyte is needed to compensate for its decomposition during formation and operation of the battery cell [24]. During the first charging cycles in the formation, some electrolyte decomposes in contact with the anode and forms the so-called solid electrolyte interphase (SEI), which acts as a passivation layer to protect the electrolyte from further decomposition [25]. Electrolyte is also consumed throughout cell operation due to the continuous ageing of the cells. For these reasons, the cells are filled with a surplus up to 40 Vol.-% [17]. Secondly, the duration of the filling sub-process depends on the ratio of the dead volume to the pore volume as well as the type of housing. With rigid housings, as with cylindrical cells or prismatic cells, the dead volume is fixed. Using flexible housings (pouch cells), the elasticity of the housing can be used to temporarily increase the dead volume of the cell. If the dead volume is smaller than the desired electrolyte quantity or the filling quantity per dispensing stroke is limited, it is necessary to fill the total electrolyte quantity in increments. Such multi-stage dosing

may result in waiting times between the dosing increments of up to more than 10 min. The waiting time is determined by the time required to ensure that enough electrolyte has penetrated into the cell body after each filling step to provide enough dead volume for the following filling step [8,26]. This in turn is based on a number of factors such as the wettability of the materials, the accessibility of the cell body and ambient conditions. Standard electrodes such as NMC and graphite electrodes, especially if calendered beyond an ideal material-specific level, have poor wettability indicated by a low wetting rate or low surface energy of the material, as do the polyolefin separators [9,27]. Approaches to improve the wettability of the materials include adaption of the porosity [27], using additives in the electrolyte, coating the separator [16] or plasma activation of the materials [22]. Regarding the accessibility of the cell body, the type of assembly is decisive. The available wetting directions are determined by the arrangement of the impermeable current collectors. With stacks, four of the six sides of the cell body are available for wetting (cf. Figure 2a), while windings only have two wetting directions (cf. Figure 2b) [26]. Additionally used materials such as insulation or tapes can restrict accessibility [28,29] (cf. Figure 2c) which can be a challenge for wetting in between filling steps and soaking later.

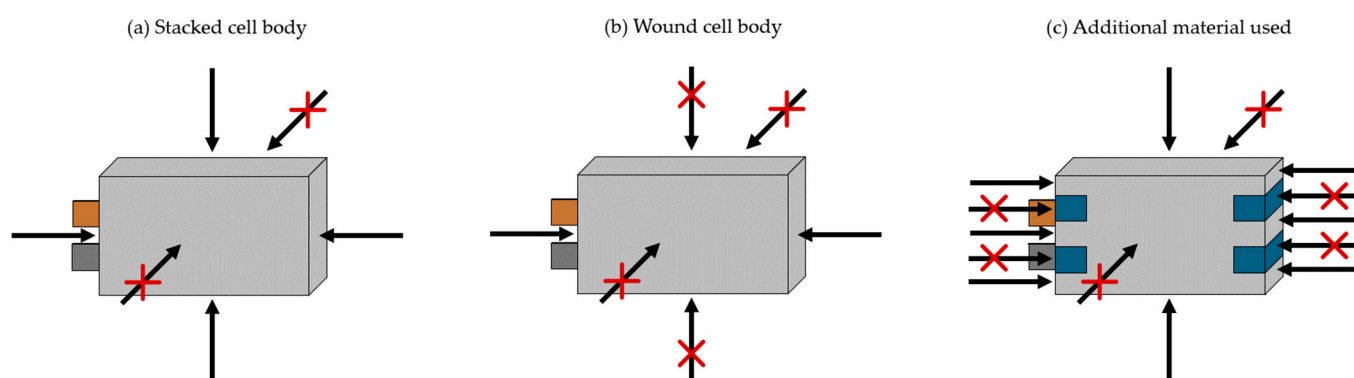


Figure 2. Available wetting directions (Indicated with an arrow) for a stacked cell body (a), a wound cell body (b) and if additional material such as tapes (Exemplary positioning) is used (c). Unavailable wetting directions are marked with a red X.

Changing the ambient conditions during electrolyte filling is associated with improving wetting and therefore decreasing the waiting time between filling steps. Changes that improve wetting are evacuation of the cell before the filling sub-process, filling with a high dosing pressure and an increased temperature level [30]. In summary, the cell formats can be categorized as follows regarding the duration of their filling sub-process. Small battery cells and cells with a flexible cell housing can be filled within seconds to minutes. Large battery cells and cells with a rigid housing take up to one hour for filling. Finally, the technical implementation of the electrolyte filling has an effect on the duration of the filling sub-process.

In the current state-of-the-art filling sub-process, the electrolyte is pumped into the cell through a filling opening by means of a dosing lance or a pressed-on dosing nozzle (cf. Figure 3).

In case of a pouch cell, a dosing lance is inserted into the open side of the pouch bag. Both types, a dosing lance or a pressed-on dosing nozzle, can be used for hardcase cells. In case of prismatic cells, a small hole in the lid assembly is used for filling, while in case of cylindrical cells, the entire can is left open for filling [12,13].

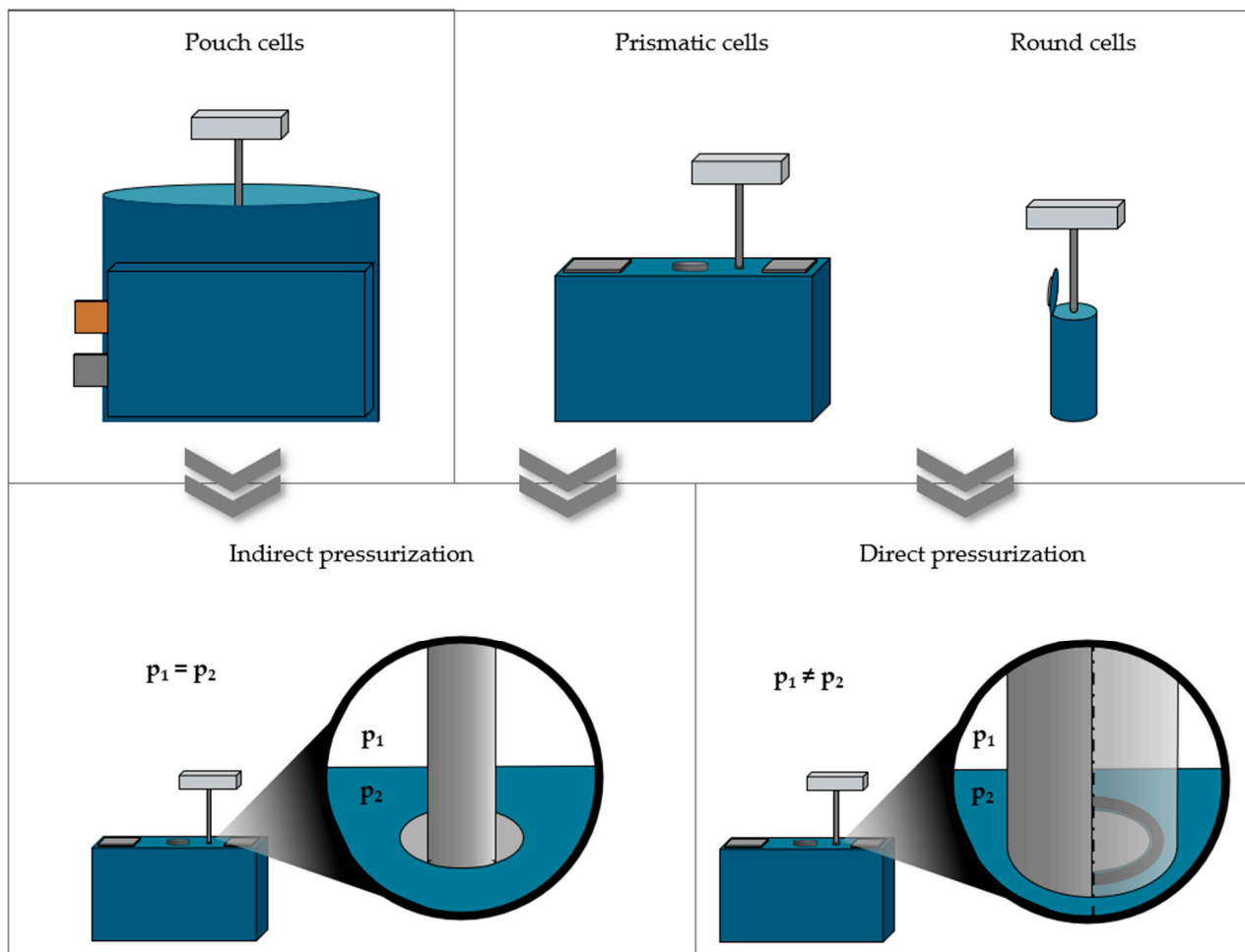


Figure 3. State of the art of filling processes for common cell formats and the corresponding type of pressurization. p_1 stands for the pressure outside the cell and p_2 for the pressure inside the cell.

Pressure can be applied to the battery cell in two ways: Indirect or direct. Both types of pressurization are shown in Figure 3. With indirect pressurization, the cell is placed inside a vacuum chamber. The atmosphere of the battery cell and the chamber are connected to each other. By varying the pressure in the whole chamber, the pressure is applied to the battery cell. With direct pressurization, the pressure is applied directly to the respective cells which is achieved with a tightly connected dosing system, such as a pressed-on dosing nozzle. Usage of direct pressurization is currently limited to hard case cells whereas indirect pressurization can be applied for both rigid and flexible housings. Direct pressurization is associated with more complex systems compared to the dosing lances, as on the one hand there is a high demand for tightness and on the other hand the dosing and pressurization need to be integrated together, for example in a pressed-on dosing nozzle. Using indirect pressurization, dosing and pressurization are carried out by two different and spatially distant systems. The main advantage of direct pressurization is the smaller volume that needs to be influenced. Instead of a larger vacuum chamber, the pressure changes only take place in the internal volume of the cell thus reducing energy consumption and saving time during pressure changes.

During electrolyte filling, the cell is commonly evacuated to pressures of around 50 mbar prior to the filling sub-process [13,31–33] with the aim of removing gas from the pores of the cell stack and thus facilitating wetting, [9,34] as well as removing residual moisture, which prevents the electrolyte from forming hydrofluoric (HF) acid solution when in contact with water [30,35,36]. The dynamic variation of the cell pressure in the range

from below atmospheric (50 mbar absolute) to above atmospheric pressure (6 bar absolute) during and after dosing of the electrolyte quantity is another typical feature [37]. These so-called pressure cycles are run with inert gases such as nitrogen to prevent side reactions with oxygen [13,38]. The purpose of the pressure cycles is to induce a transport movement of the free liquid electrolyte which influences the wetting positively [26]. Although the low pressures during filling are advantageous for the distribution of the electrolyte, they also have a few drawbacks. Due to the low pressures, evaporation increases. The composition of the electrolyte changes over time due to the varying volatility of the electrolyte components. The evaporation affects the properties and the amount of the electrolyte in the battery cell. In addition, cavitation may occur in the filling system if vacuum conditions prevail there as well [9]. However, as the acceleration of the wetting by the vacuum is essential, an optimum pressure must be found at which the disadvantages mentioned are kept within limits.

In addition to the implementation of pressurization, the automation of the process plays an important role. Both manual filling and semi-automatic filling [39,40] of the battery cells are used on a laboratory and pilot scale which means that the cells are produced in batches. In large-scale production of battery cells, fully automatic filling systems [41,42] that work continuously are usually used to reduce the process costs per unit produced.

Overall, the state of the art seems to have hardly changed for several years [13]. Admittedly the scientific knowledge base regarding electrolyte filling, especially focused on the wetting sub-process, has expanded. However, the divergence of the specific process design and technical implementation of the filling sub-process remain which is partly caused by a lack of scientific studies on the filling sub-process at industry-relevant scale. The main challenges are still a process time reduction while ensuring a high-quality product with homogeneous wetting, high filling accuracy as well as high process safety and stability [43].

3. Methods

The implementation of novel electrolyte filling technologies has the potential to optimize the process, thereby addressing the main challenges associated with the filling sub-process previously described. Therefore, we aim to analyze and evaluate novel technologies for electrolyte filling. We first conducted a comprehensive literature search to identify technologies that could be implemented as a result of the transfer of knowledge and technology from the scientific sector to industry. The literature search included peer-reviewed scientific journal articles and conference papers and was done with Scopus, which is the largest database for peer-reviewed literature [44]. Using the following query, the literature search was conducted in September 2023:

TITLE-ABS-KEY ("Electrolyte filling" AND "Lithium-Ion battery").

In order to include technologies originating from industrial research and development, a patent search was conducted to identify any relevant novel technologies associated with the process of electrolyte filling. Patents offer a valuable source of insight into the initial phases of technological advancement. They are a commonly utilized metric for technology forecasting, as they are indicative of emerging technological trends [45]. Patbase was selected as a database for the patent search done in September 2023 because it is referred to as "the industry leading global patent database" [46]. The search query below was utilized:

TAC = ((Electrolyte Filling Device OR Electrolyte Filling Equipment OR Electrolyte Injection Device OR Electrolyte Automatic Filling System OR Liquid Filling Device OR Injecting Electrolyte OR Method for filling electrochemical cells) AND (lithium OR (Li-Ion OR lithium-ion))) AND CPC = (H01M50/60 OR H01M10/052 OR H01M10/058)

The literature search returned 43 hits while the patent search yielded 87 hits. To facilitate the targeted processing of these records, the data set was analyzed in three steps (see Figure 4).

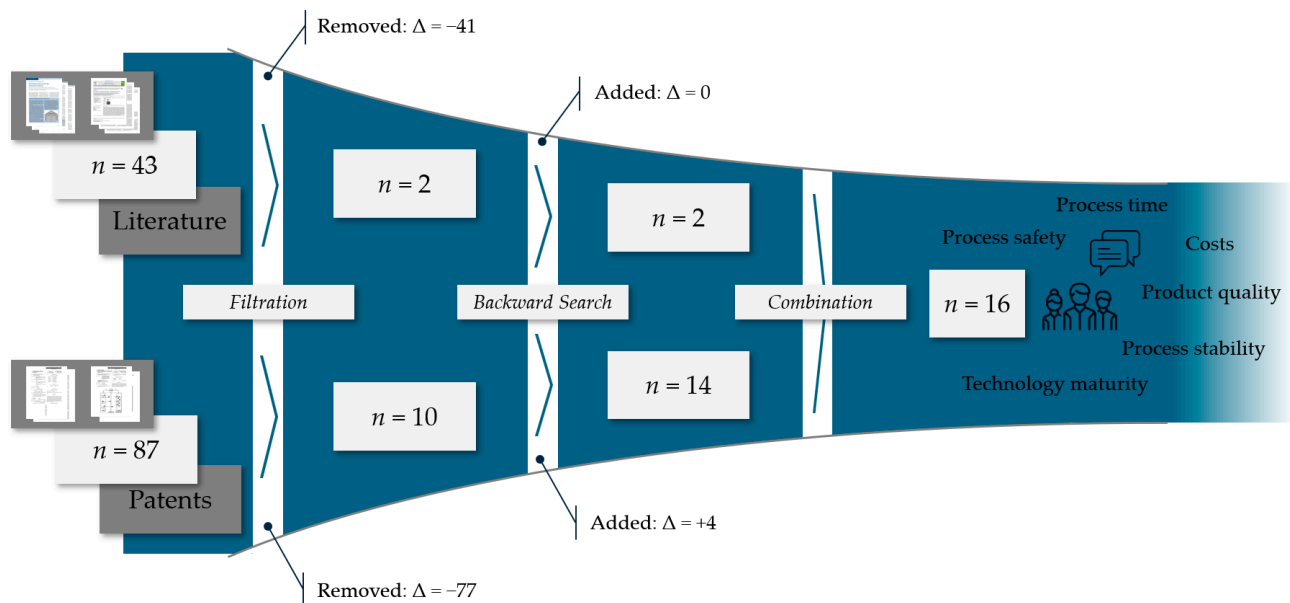


Figure 4. Overview about the publication and patent selection process.

In the first step, all publications and patents that were inaccessible or did not present novel technologies for electrolyte filling were excluded. By novel electrolyte filling technologies, we mean technologies that deviate from the state of the art as described in Section 2 and that are used in the filling sub-process as defined above. In the second step, additional relevant publications and patents were identified through a backward search of the sources of the remaining records. Subsequently, the data set was merged into one comprehensive set. As a result of this analysis, the records were narrowed down to $n = 16$ relevant publications and patents, which happen to contain sixteen distinct technologies.

In the next step, the data collected on the sixteen technologies was aggregated into a matrix for comparison of the technologies. A group of experts, consisting of three researchers from the fields of technology management, process engineering and mechanical engineering, then evaluated the technologies based on the criteria of technology maturity, respectively technology readiness level (TRL), costs, process time, process safety, process stability and product quality. The evaluation criteria are based on the main challenges outlined in Section 2 and incorporate additional considerations such as costs and TRL to contextualize purely technical criteria. Furthermore, the criteria are consistent with those set forth in relevant publications in the domain of technology benchmarking [47,48]. In order to ensure the objectivity of the analysis, the rating steps were conducted independently by all experts and critically discussed afterward. The discussion was characterised by a high degree of iteration, whereby the evaluations of technologies recorded subsequently were compared with those recorded previously. Based on the five individual criteria, a superordinate evaluation was made, which is referred to as “overall evaluation” used to identify the most promising technologies among those evaluated. With the exception of the TRL, all individual criteria and the “overall evaluation” were evaluated using the following scale:

- +++ Much higher potential than the reference
- ++ Higher potential than the reference
- + Slightly higher potential than the reference

- 0 Same potential than reference
- Slightly higher potential than the reference
- Lower potential than the reference
- Much lower potential than the reference

The TRL can be used as a quantitative indicator to assess the level of technological development. It employs a scale ranging from TRL 1 to TRL 9. This study classified each technology according to the TRL definitions from the National Aeronautics and Space Administration (NASA) [49] and the Deutsches Institut für Normung (DIN) [50].

4. Results and Discussion

Almost all the sixteen technologies evaluated were found in patents, indicating that the development of the technologies originates primarily from industrial players and less from the scientific community. Many of the scientific publications explore theoretical background of the wetting sub-process and the resulting practical implications on the overall electrolyte filling process.

In this chapter, all technologies are first described and discussed based on the selected criteria (see Section 3). The five most promising technologies are then highlighted and compared in more detail. It should be noted that not all the sixteen technologies are in competition with each other and are mutually exclusive. In some cases, they can complement each other or in other cases are limited to certain cell formats or applications.

4.1. Pressurization up to 10–20 Bar

Pressurization of battery cells is common in the context of pressure cycles (see Section 2). The two property rights CN108832071 B and CN108054337 A deal with a filling method for hard case cells and in this context specify the extent of pressurization to a pressure range of 3–8 bar (CN108832071 B [33]) and 5–10 bar (CN108054337 A [51]) respectively. The company IP Power Systems GmbH advertises a pressure range of up to 6 bar absolute for their system technology [52]. The overpressure reduces the process time due to faster injection and delivery of the wetting agent [53]. A further reduction in process time is assumed for pressurization between 10 and 20 bar. In case of direct pressurization, mechanical stability of the cell housing limits the applicable pressures. For example, the bursting membrane of a prismatic cell can withstand a pressure difference of 7.8 to 8.8 bar [54]. Applying an external pressure or changing to indirect pressurization would be necessary. Higher pressures also make damage to the cell stack more likely and process control and precision more difficult, so that process stability and product quality are impacted negatively. The higher the pressure, the riskier the process becomes for the operator and material goods, consequently the demands on the system and therefore the costs increase significantly.

4.2. Pressurization up to 20–30 Bar

Although pressurization to 20 to 30 bar may further reduce the process time compared to 10 to 20 bar, it is accompanied by considerable uncertainties. On the one hand, there are no reports of the use of such high pressure, so there are doubts as to whether the process time is reduced to the same extent as for lower overpressure ranges or whether a diminishing marginal utility is reached. On the other hand, the costs and risks increase with a further increase in pressure due to higher demands on the system. Overall, a poorer evaluation compared to the reference is attested.

4.3. Multiple Filling Points

This technology covers all applications in which the electrolyte is filled into the battery cell at several points (simultaneously), e.g., the use of several dosing lances across

the entire length of a pouch bag. Although this technology is mentioned in scientific publications [11,26], it has not yet been patented. The benefit of filling at several points becomes larger the larger the cells become, as process time can be saved through faster dosing of the required electrolyte amount. One of the main disadvantages of this attempt are the costs associated with the parallel multiplication of system parts. If changes are made to the cell design, they will also lead to higher costs in upstream and downstream processes. Overall, the costs will exceed the benefits of this technology.

4.4. Screw Connection

The screw connection can be implemented as a screw hole in the cover assembly of a cell, as described in IP CN202076337 U [55], or as a protruding thread on the cell, as seen in IP Power Systems [52]. The application of a screw connection is limited to hard case cells, whereby the port technology is the equivalent for the pouch cells. By using a screw connection, direct pressurization can be used conveniently, which saves costs in operation compared to indirect pressurization, as the reduced volume leads to lower energy costs. There are also advantages in terms of safety and stability of the process, as the screw connection is tight, firm, and industrially established. On its own, the use of a screw connection slows down the filling sub-process, but this technology is an enabler technology for the use of extreme pressurization. For pressures of 10–30 bar, an O-ring seal will not be sufficient.

4.5. Large Filling Opening with Burst Diaphragm

The technology is described in property right DE102022001429 A1 [56]. It is intended to be used for prismatic cells. The electrolyte is not filled in through a small additional hole in the cover assembly of the cell, but through a large opening into which the bursting membrane is later welded. Due to the larger opening, the gases can escape more quickly, and the electrolyte can be filled in more quickly, thus reducing the process time. The extent of these savings is not quantified in the property right DE102022001429 A1. By using the opening for more than one function, the additional hole and sealing parts and processes can be saved. The necessary changes to the cell design would initially lead to higher costs due to the deviations from the standard cell. Possible cost savings would only arise in the long term with large-scale production. The use of a large filling opening with burst diaphragm has no impact on the dimensions of process reliability, process stability and product quality compared to the state of the art. Overall, it is unclear whether the reduction in process time as an advantage outweighs the disadvantages of deviating from the standard cell design and the risk associated with welding the bursting membrane.

4.6. Port Technology

In so-called port technology (see property right DE102019003594 A1 [57]), which can be used for pouch cells, access points (ports) are inserted into the pouch film. While the inside of the cell is encapsulated from the environment, operations such as filling, evacuation, direct pressurization or degassing are possible through the ports. In contrast to most of the other technologies described in Section 4, it has been further developed and is already being marketed by IP Power Systems GmbH [52]. By enabling direct pressurization of pouch cells, costs and process time can be saved as less volume needs to be pressurized or evacuated. In addition, the process time can be reduced, as the port technology also enables parallel filling or simultaneous execution of different process steps at several points in the pouch bag. In terms of process safety, the technology is advantageous as the system is sealed and thus prevents contamination of the periphery or the cell.

4.7. Flushing of the Cell by Electrolyte Pump Circuit

Information on this technology is known from property right CN110416485 A [58]. The battery cell is flushed with the electrolyte, which is pumped in a circuit. Pressure cycles are not used. On the one hand, product quality is improved by avoiding pressure induced deformation of the cell housing and flushing particles and impurities out of the cell. On the other hand, the stronger flow in the cell can negatively affect the product quality by displacing components or causing damage due to entrained impurities. A significant acceleration of wetting, as postulated in the property right, would reduce process time by an unquantified amount, but due to major deviations from the state of the art, the costs of the process increase. The cell design and the filling system would need to be modified to implement the technology. In addition, the electrolyte takes particles with it during circulation, which is why it must be treated to remove them. Overall, the experts assess the technology as negative compared to the reference.

4.8. Vibration of the Dosing Lance

The vibration of the dosing lance comes with minor changes to the state of the art. As described in patent CN113314813 B [59], the vibration can be generated by the contact between triangular plates and a side support when the dosing lance is raised. This technology reduces dripping and thus contamination and corrosion on the periphery and the cell. The slight increase in system complexity is associated with costs. Otherwise, the technology has no impact on the evaluated dimensions. Overall, a positive evaluation compared to the reference is attested.

4.9. Telescopic Plates in the Injection Tube

Like the vibration of the dosing lance, the so-called telescopic plates also reduce contamination on the periphery and the cell. One or more telescopic plates are placed in the dosing lance, which close and seal the dosing lance after electrolyte dosing. As a result, electrolyte dripping is prevented (see property right CN115000649 B [10]). This technology slightly increases costs but is advantageous in terms of process safety, as it prevents contamination of system parts or battery cells and thus reduces the potential for hydrogen fluoride (HF) formation. In contrast to vibration, the mechanics are in the dosing lance itself and therefore have a negative effect on process stability, as impurities can accumulate.

4.10. Telescopic Capsule in the Injection Tube

Like the telescopic plates and the vibration, the telescopic capsule is designed to reduce contamination. The mode of operation is like that of the telescopic plates. According to property right CN114400422 A [60], the dosing lance contains an expandable bladder whose atmosphere is connected to components outside the lance. By moving these components, the expansion of the bladder inside and thus the flow of the electrolyte in the dosing lance is precisely controlled which improves process stability at the expense of costs. Except for process stability, the telescopic capsule is rated the same as the telescopic plates due to their similar mode of operation. Since this technology is less likely to be affected by accumulated impurities, it has a better rating regarding process stability.

4.11. Flexibilization to Different Cell Dimensions

Flexibilization to different cell dimensions refers to a group of technologies that are used to fill cells of varying dimensions in one filling system. They can be used not only instead of, but also in combination with the other technologies discussed in this study. Flexibility regarding different cell dimensions is mentioned in many patents [61–64]. In

concrete terms, flexibilization is implemented, for example, by using height-adjustable dosing lances or injection heads (see property right CN109192920 A [61] or CN110311085 B [62]) in order to enable the filling of cells of different heights. Patents also contain devices in which cell holders or conveyor belt side guides are spring-loaded to be able to hold cells of different widths and depths within a certain flexibility corridor (see property right CN111403677 A [63] and property right CN110311085 B [62]). The flexibilization regarding different cell dimensions does not affect the process time of the electrolyte filling but generates additional costs upfront. In addition, process safety and process stability suffer from such flexibilization, as movable or flexible components can diminish repeatability and reliability. Flexibilization would be advantageous if frequent changes to the cell design are expected. As frequent changes to the cell design are not to be expected in the context of a gigafactory, this technology was assessed as negative overall.

4.12. Filling by Centrifugal Force

Filling by centrifugal force differs significantly from the state of the art electrolyte filling as the force applied to fill the electrolyte into the cell is not provided by a pump. According to property right CN209607835 U [65], a large number of battery cells are placed in cell holders, which in turn are placed in a centrifuge. The electrolyte is pressed into the cells by the rotational movement of the centrifuge intended to prevent the electrolyte from evaporating, as no vacuum is required. This procedure may lead to an accelerated distribution of the electrolyte, although this claim is not verifiable due to the scarcity of information. It should be noted that process reliability of the filling sub-process decreases in comparison to the state of the art because the centrifugal forces affect all parts of the cell, including the cell stack, potentially leading to displacement of and damage to the cell stack.

4.13. Electrolyte Spraying

Electrolyte spraying refers to an electrolyte dosing unit for cylindrical cells. Unlike dosing lances, it closes directly with the opening of the cells and sprays the electrolyte into them via many nozzles. This fine dispersion of the electrolyte is intended to avoid high surface tension between electrodes and separator, which makes electrolyte filling more difficult. The technology is described in property right CN106025168 B [59]. As a result of the fine distribution, it is possible to shorten the process time by an amount not quantified in the property right. At the same time, the high number of nozzles and fine distribution in droplets leads to an increased risk of contamination on the periphery and cell, which reduces process reliability.

4.14. Filtration

Filtration is an established technology in other sectors, such as the chemical industry, and is certainly used in some electrolyte filling systems (e.g., see property right CN109473621 B [66]). Filtration is a mechanical process in which solid particles are separated from a liquid or gas using a porous filter medium. The fluid flows through the filter medium driven by a pressure difference and is thus freed from particles [67]. With filtration, the process time and the costs will increase. However, by removing impurities, undesirable effects during the process, such as clogging of the tubes, or during operation of the battery cell, such as short circuits [68], are avoided and thus product quality is improved. It also acts as an enabler for other technologies such as high pressurization, high temperatures and other innovation approaches. The build-up of a filter cake will change filtration performance and pressure behavior over time leading to negative impacts on the process stability.

4.15. Just-in-Time Mixing

With just-in-time mixing, the various components of the electrolyte are only mixed on site in the electrolyte filling machine instead of dosing the finished electrolyte directly into the cell. This technology is described in property right DE102020132021 A1 [69]. It prevents the electrolyte from decomposing and reacting during transportation or storage. Just-in-time mixing leads to the optimal composition of the electrolyte, which achieve optimum product properties. As a result, the product quality in terms of maximizing cell performance can be improved compared to the state of the art. However, increasing the complexity and number of system components has a negative impact on process reliability and costs. Overall, the suitability of the technology depends on the use case. The technology can prove advantageous for smaller production lines, as electrolyte dosing is made more flexible with just-in-time mixing. For gigafactories, it is rated as negative, as liability for errors and warranty claims arise for the cell manufacturer.

4.16. Collecting and Recycling

The filling device described in property right CN113314813 B [70] also mentions a collecting basin. The basin is placed under the battery cell or electrolyte dosing unit and allows the collection and recycling of spilled electrolyte. Contamination can be contained in the event of spillage and thus process reliability can be increased. However, the priority should be to avoid spillage during electrolyte filling, so this technology is only suitable to a limited extent for systems that exhibit significant dripping or overflowing during filling. Collecting and treating the spilled electrolyte add to the costs, making this technology not worthwhile for a cleanly running filling system.

4.17. Summary

The technologies described are summarized in Table 1 along with the corresponding results of the expert evaluation based on the criteria of technology maturity (TRL), costs, process time, process safety, process stability and product quality (see Section 3 for the legend of the evaluation scale). An overarching evaluation for the novel technologies can be seen in the last column. Furthermore, the technologies were also divided into the groups: *process parameter*, *cell design*, *filling device* and *electrolyte* to ensure a better overview and to roughly categorize them.

Table 1. List of novel technologies and the corresponding expert assessment.

Group	Technology	TRL	Cost	Process Time	Process Safety	Process Stability	Product Quality	Overall Evaluation
Process parameter	Pressurization to 10–20 bar	4	--	++	--	-	-	++
	Pressurization to 20–30 bar	2	---	+++	---	--	--	+
Cell design	Multiple filling points	5	--	+	0	-	0	-
	Screw connection	7	++	-	++	+	0	+
	Large filling opening with burst diaphragm	3–4	0	+	0	0	0	0
	Port technology	7	+++	++	++	-	0	+
	Flushing of the cell by electrolyte pump circuit	3	---	+++	-	--	0	--
Filling device	Vibration of the dosing lance	3	-	0	+	0	0	+
	Telescopic plates in the injection tube	3	-	0	+	--	0	0
	Telescopic capsule in the injection tube	3	-	0	+	-	0	0
	Flexibilization to different cell dimensions	3	--	0	-	-	0	-
	Filling by centrifugal force	3	-	+	-	0	0	-
	Electrolyte spraying	3	-	+	-	-	0	-
Electrolyte	Filtration	8	--	0	++	--	+	0
	Just-in-time mixing	4	--	0	0	-	++	0
	Collecting and recycling	3	--	0	+	0	0	-

To provide a better overview, the technologies are visualized in a portfolio analysis in Figure 5 based on their overall evaluation (cf. Table 1) and technology maturity. In addition, the color of the data points shows which cell formats the respective technology is limited to. It is noticeable that almost all the technologies examined are suitable for hardcase cells (cylindrical, prismatic). Just over half of the technologies are available for pouch cells. One possible reason could be the prevailing opinion that pouch cells are easier and quicker to fill due to their flexible housing. As a result, less development effort is attributed to the filling concept of pouch cells. Port technology contradicts this trend, as it was developed exclusively for pouch cells. The heat map also shows that many of the technologies are classified on a TRL of 3 to 4 in combination with a slightly negative (-) or neutral evaluation (0). On the one hand, this recurring classification results from this paper's objective of identifying novel technologies that generally have a low technological maturity and no visible significant advantage over the state of the art. On the other hand, the assessment must be continuously adapted as the technologies evolve and mature.

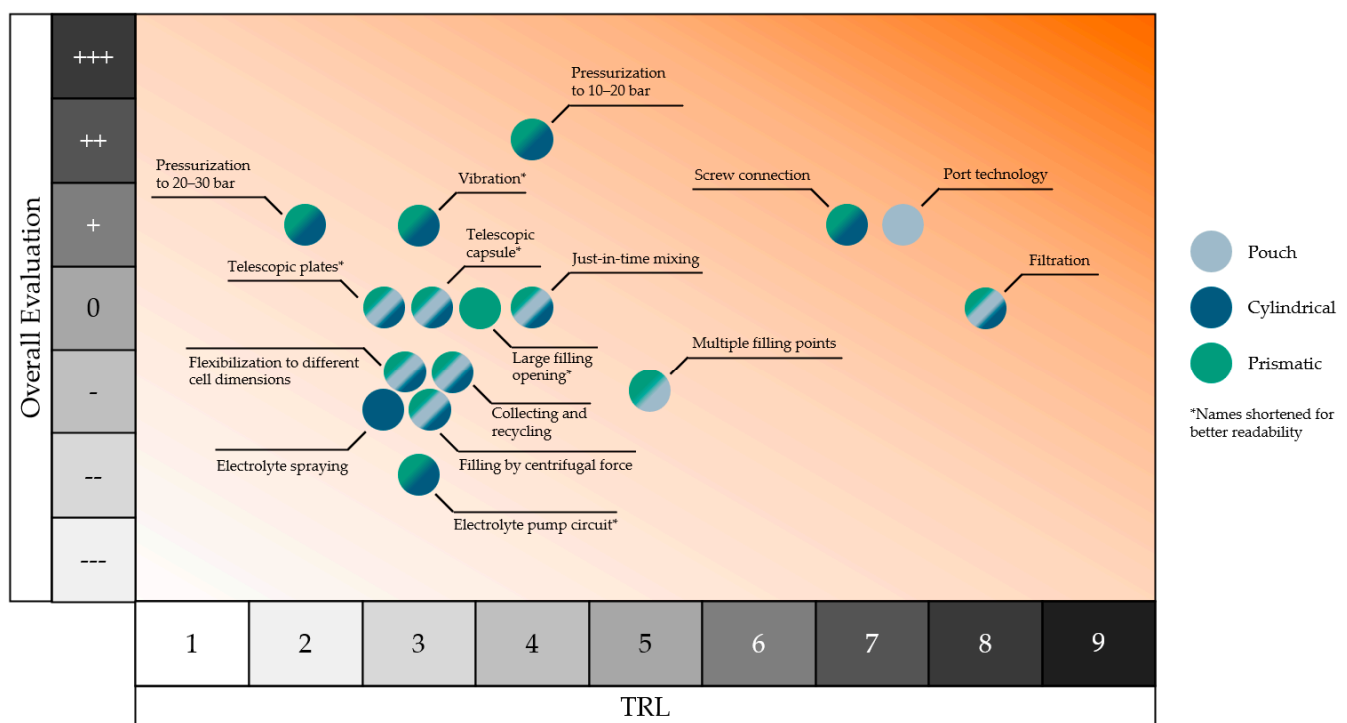


Figure 5. Overall evaluation (y-axis) of all technologies depending on their technological maturity (x-axis) including their area of application regarding cell format.

There are a total of five technologies that have received a positive assessment compared to the state of the art and thus have the best overall evaluation. They are also highlighted in Table 1. These include pressurization to 10–20 bar and 20–30 bar, the screw connection, port technology and vibrating the dosing lance. Pressurization can only be rated positively for the process time criterion, but pressure is of paramount importance there. If indirect pressurization is used, the technologies can be used for all cell formats. Direct pressurization, on the other hand, can be readily implemented only with hardcase cells, whether cylindrical or prismatic. Pressurization has a low TRL, whether for 10–20 bar (TRL 4) or 20–30 bar (TRL 2). However, as pressurization up to a maximum of 10 bar is already state of the art, a development towards higher pressures and thus a further development of these technologies is expected.

The screw connection might also be beneficial for this development. It can be seen as an enabler technology for the use of higher pressures. It can be used with hardcase

cells and offers a more secure connection between the filling system and the cell, which is essential at higher pressures. In addition, the screw connection itself offers the advantage that it effectively prevents the cell from overflowing. Screw connections are widely used in industry and occasionally in electrolyte filling systems, so that a TRL of 7 is reported for this technology. The port technology has a similar maturity. Although it can only be used for pouch cells, it offers the advantage of allowing direct pressurization for them, which is not possible without the port technology. The technology has a positive effect on costs, process time and process reliability, making it one of the promising technologies overall. In contrast, the vibration of the dosing lance hardly changes the electrolyte filling process. However, it increases process reliability, as the dripping of electrolyte from the dosing lance is controlled. At the same time, the costs increase only slightly, making it advantageous overall. Furthermore, the technology does not exclude any others, but can rather be used in combination with other technologies. With a TRL of 3, this technology still requires significant development. The evaluation of the technologies with an overall evaluation > 0 along all five dimensions is illustrated in Figure 6.

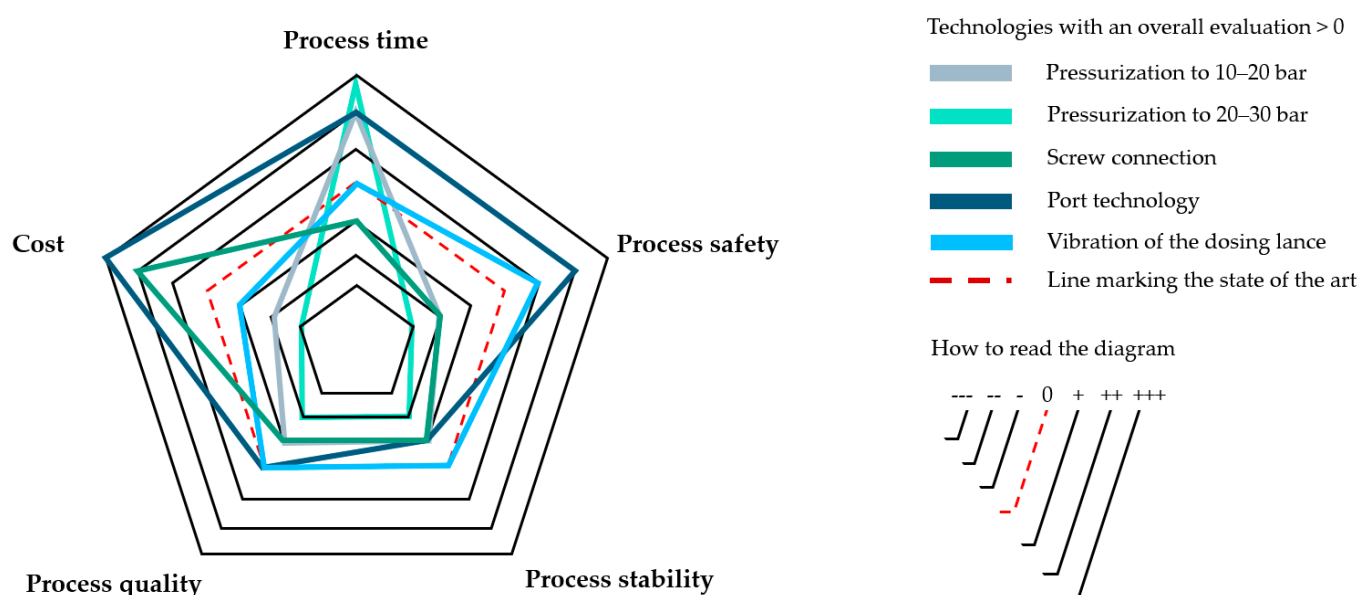


Figure 6. Radar chart illustrating the detailed assessment of the technologies with an overall evaluation > 0.

During the research, it was generally noticeable that the availability of information on the technologies is very limited. Most of the information comes from patents that do not contain any quantitative data for assessing the evaluation criteria. Moreover, the majority of the assessed technologies are not implemented in industry, which is reflected in a low TRL. Considering their neutral to poor assessment compared to the state of the art, many of the technologies will not make it to market maturity. Except for vibrating the dosing lance, the promising technologies are all aimed at modifications regarding pressurization. It is to be expected that direct pressurization will become increasingly important and that this trend will be accompanied by increasing pressure. This assessment also shows that no disruptive technology for the electrolyte filling sub-process is discernible. Mainly incremental improvements regarding costs, process time, process safety, process stability or product quality to the process can therefore be expected in the coming years. Significant savings in process time can be expected when pressurization with a pressure of up to 30 bar is ready for the market.

5. Conclusions

The available literature on electrolyte filling is largely confined to the incremental acquisition of knowledge about the processes occurring in small cell formats within a laboratory setting. It is notable that studies related to industrial applications and focusing on large cell formats are scarce. Nevertheless, they are of significant value in providing insight into scaling effects and process optimization for industrially applicable cell formats. Consequently, they are essential for the optimization of the electrolyte filling. This review offers a valuable contribution to the existing literature by providing a comprehensive synthesis of the limited and fragmented information on this topic, making it readily accessible within the broader academic landscape. To this end, a systematic literature and patent review was conducted to identify novel electrolyte filling technologies. Sixteen relevant technologies were identified and subsequently evaluated by a group of experts from the field of technology management, process engineering and mechanical engineering. The criteria for the evaluation were technology maturity (TRL), costs, process time, process safety, process stability and product quality as well as a superordinate evaluation, the “impact on problem solving”. Five promising technologies were identified, including pressurization to 10–20 bar and 20–30 bar, the screw connection, port technology and vibrating the dosing lance, all of which were positively evaluated compared to the current state of the art. Pressurization is highly beneficial for reducing process time whereby indirect pressurization can be used for all cell formats and direct pressurization only for hardcase cells. As pressurization up to 10 bar is already common, further development towards higher pressures is anticipated. The screw connection is required for handling higher pressures, providing a secure connection that prevents electrolyte spillage. Port technology enables direct pressurization of pouch cells, positively impacting costs, process time, and reliability. Lastly, vibrating the dosing lance minimally changes the filling sub-process but increases process safety by controlling electrolyte dripping, with only slight cost increases. Except for vibrating the dosing lance, all promising technologies focus on pressurization modifications. However, this does not apply to many of the other technologies. Overall, these predominantly have a low TRL of 3 to 4 and a slightly negative (-) to neutral rating (0). As it is difficult to estimate future developments due to the low level of technological maturity and limited availability of information, the assessment must be continuously adapted as the technologies evolve and mature.

The findings of our study have a number of practical implications for stakeholders from the realms of research, industry, and politics. The research community is made aware of promising research fields, while industry players are informed of potential (alternative) technologies through this review and are thus able to compare them with their own R&D activities. Furthermore, funding managers are supported by the results in their decision-making regarding which investments to promote and which topics are more promising for successful technological breakthroughs than others.

This review is limited in two ways: in terms of its methodology and in terms of the content it covers. The methodology of this study relies on largely qualitative information to perform a quantitative assessment of technologies. Despite our best efforts to consider and evaluate all novel electrolyte filling technologies in an objective manner, it is not possible to guarantee with absolute certainty the completeness of the information base. This may be due to the fact that some technologies are treated as industry secrets or because it is generally challenging to obtain information about novel technologies. As the scope of this study is the filling sub-process, technologies related to the wetting sub-process were not considered. In addition, this study dealt with tangible technologies, so that further studies could address the potential for optimization through digitalization or simulations, for example. As a final remark, the literature and patent research showed the focus of the

scientific community on the wetting sub-process, so that the scientific investigation of the filling sub-process, especially in an industry-relevant scale, has knowledge gaps that still need to be closed.

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