

# **Economical and ecological production of Li-ion pouch cells by effective automatization and climatic engineering**

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## **Summary**

Electro mobility plays a prominent role in the German federal government's targeted energy transition. A growing market for energy storage systems, the constant development of battery cells and an awareness of sustainable production are inevitably leading to a demand for innovative, modular plant concepts which can easily be adapted to changing conditions and satisfy core requirements in terms of effectiveness, economy and ecology. Technical solutions and automated machines are currently only available to a limited extent on the market. To achieve targeted ecologically and economically attractive cell manufacturing, a review of the literature was conducted to show the significant potential along the value chain. It emerged that there is substantial potential to reduce the cost and time of production at the point when lithium-ion pouch cells are filled with electrolyte. Based on this knowledge, a filling concept was created which is realized by a plant concept consisting of two devices. With this prototype it is possible to increase the wetting speed of lithium-ion pouch cells by varying the filling position and the vacuum control in the cells before filling. Furthermore, the formation gas can be sucked directly out of the cell, making a gas bag superfluous. The production costs can be reduced and waste production decreased. By using a mini-environment, the costs of air conditioning can also be reduced, additionally easing the burden on the environment.

*Keywords: lithium battery, electrolyte, cost, environment, automated*

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## **1 Introduction**

Electromobility plays a prominent role in the German federal government's targeted Energiewende or energy transition [1]. However, innovative storage technologies also require novel production systems which are capable of producing the necessary number of pieces at the required level of quality under ecological and economical conditions.

Technical solutions and automated machines are currently only available to a limited extent on the market. A growing market for energy storage systems, the constant development of battery cells and an awareness of sustainable production is inevitably leading to a demand for innovative, modular plant concepts which can be easily adapted to changing conditions and satisfy core requirements in terms of effectiveness, economy and ecology.

The aim of this project is to solve essential scientific questions as the basis for developing a modular manufacturing and plant concept. In sum, the plant concept promises to reduce process energy, material and operating costs, leading to ecologically and at the same time economically attractive cell manufacturing. [2]

## 2 Preliminary investigations and methodology

To achieve the target of ecologically and economically attractive cell manufacturing, a review of the literature was conducted to show the significant potential along the value chain. In a second step, the publications thus found were evaluated and filtered to select the most relevant issues. To validate the core statements of the publications, different pilot tests were carried out using an experimental setup. After the evaluation of the experimental tests, the expertise gained was then directly incorporated into the development of the demonstrators.

### 2.1 Literature review

In the course of the literature review, three themes were examined. The first was the influence of the separators and the electrodes on the wetting behaviour. For this purpose the orientation of the separator, the filling position and the internal streaming of the electrolyte in the battery cell were investigated along with the impact of vacuum. The aim was to find an association between the above-mentioned contributing factors and the penetration of the active components of the battery cell by the electrolyte.

It was ascertained that the orientation of the separator in the cell depends on its manufacturing process or on the production direction. The separator is wound onto coils to ensure continuous production. Ultimately, this means that the direction can only be influenced during the stacking process. When the stack is produced, the separator's orientation can only be varied to a very limited extent when using the z-folding process. This method has particular advantages compared with stacking with single-sheet separators. For that reason and the related expected low flexibility and benefit, research in this field has been abandoned. [3]

There is much higher potential for pressure variation in the cell before, during and after the electrolyte filling because there is no dependency on previous production processes. Furthermore, the filling process does not depend on the cell dimension, allowing for greater flexibility and application maturity. It became clear that a pressure pulsation inside the cell is advantageous. The pump effect generated in this way accelerated the wetting speed of the active material, resulting in a lower wetting time. [4], [5]

Beside the influence of the pressure, the effect of varying the filling position was investigated. The literature review shows that there are no relevant publications on this topic. In order to consider this issue, a test set-up has been developed which is described in more detail in Section 2.2.

Another important subject of discussion was the economic and ecological aspects of battery cell manufacturing, and especially the influence of air conditioning. In this context, it became apparent that there is substantial potential to reduce the cost and time of production at the point when the cells are filled. The environmental impact could also be reduced significantly.

Finally, patent research was carried out to show the current state of the art and to detect potential gaps in the field of electrolyte filling. It was determined that there are no relevant patents in the context of an automated electrolyte filling device; any such patents have expired. [6], [7], [8], [9], [10], [11], [12]

### 2.2 Experimental setup and procedure for preliminary investigations

Based on the research results, an experimental device (see Figure 1) was designed to gain a deeper knowledge of pressure variation and the variation of the filling position. Furthermore, a filling concept was developed.

#### 2.2.1 Experimental setup

The experimental device consists of a frame holding the test cell. The frame contains four ports to inject the electrolyte or create a vacuum in the cell. The seal on the connection between the port and test cell is guaranteed by sealing the needles. To enable injection and suction, there are four pneumatically actuated syringes. They can be regulated by four pressure controllers. In addition, a manometer was attached to one of the connections to measure the pressure in the test cell. The liquid-carrying pipes and valves are made of polytetrafluoroethylene (PTFE) and polypropylene (PP) as they are resistant to solvents.

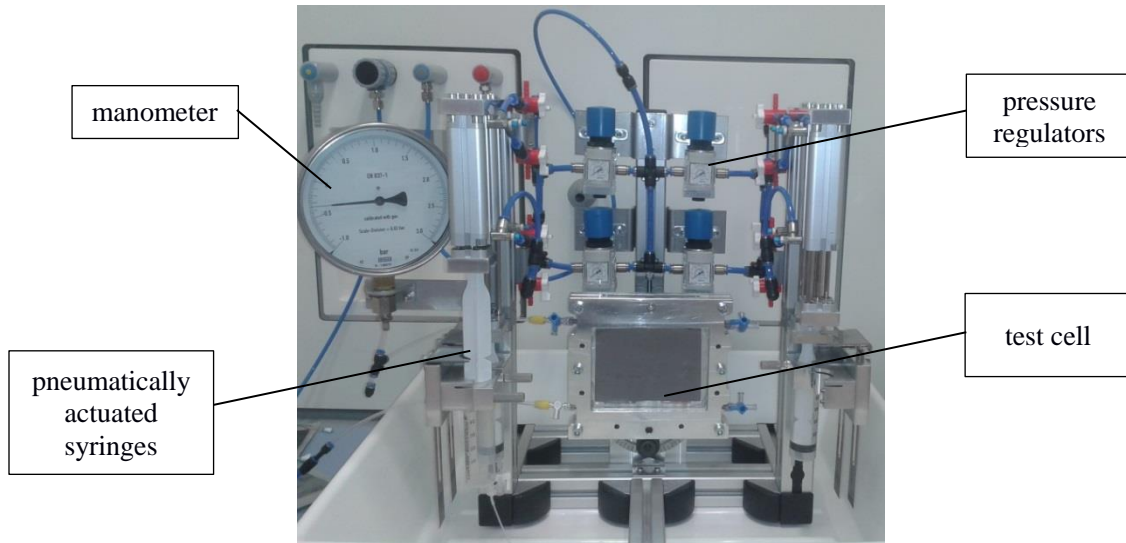


Figure 1: Experimental setup for preliminary investigations

### 2.2.2 Experimental procedure

To analyse the flow of the electrolyte in the cell and the effect of varying the injection and suction position, four test scenarios were developed. In this context, a diagonal stream, a linear stream at the bottom side and a diagonal and linear stream were investigated at the same time (see Table 1). Before filling, the cells were evacuated to a pressure of  $p_{eva}=150\text{mbar}$  (abs).

Table1: Schematic representation of the experimental procedure

Test	schematic representation of the injection and suction position
Test 1: injection at top right & suction bottom left	
Test 2: injection at bottom right & suction bottom left	
Test 3: injection on left & suction at bottom right	
Test 4: injection at top & suction at bottom	

In every test the filling volume was 18 ml of electrolyte, which entered the cell with a volume flow of 1.2ml/s. To be able to perform a temporally discretised evaluation, the tests were filmed with a camera. The wetting of the cell in the various experiments was compared by optical analysis. The comparison took place 2, 5, 10, 30 and 60 seconds after the filling started.

### 2.2.3 Experiment evaluation

During all tests, except Test 4 “injection at top & suction at bottom”, a dry spot remains in the upper area of the cell up to a time of 60 seconds (red mark; see Figure 2 c)). The worst wetting is observed during Test 2 – “injection at bottom right & suction bottom left”. In this case the wetting is only supported by the capillary effect.

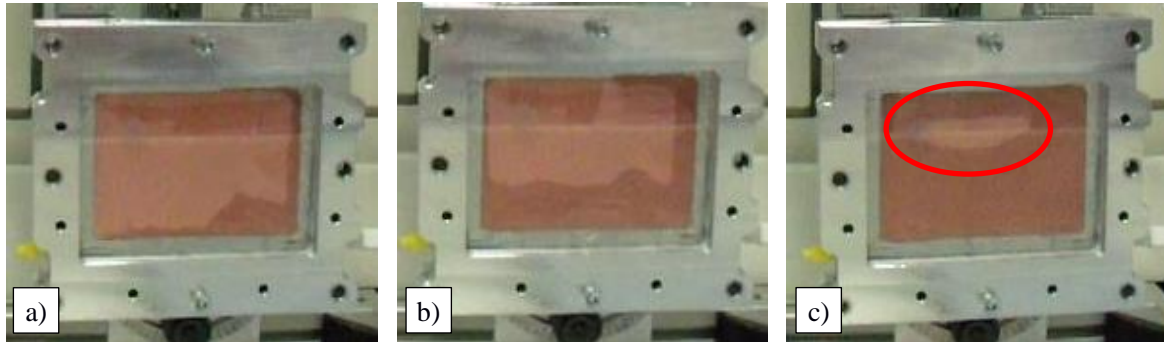


Figure 2: Test 2 – Wetting after a) 5s; b) 10s; c) 60s

The best results were obtained during Test 4 (see Figure 3). After 10 seconds all areas were wetted. It is presumed that the wetting is forced by gravity as well as the capillary effect. The diagonal flow in the cell also has a positive impact on the distribution of the electrolyte in the cell.

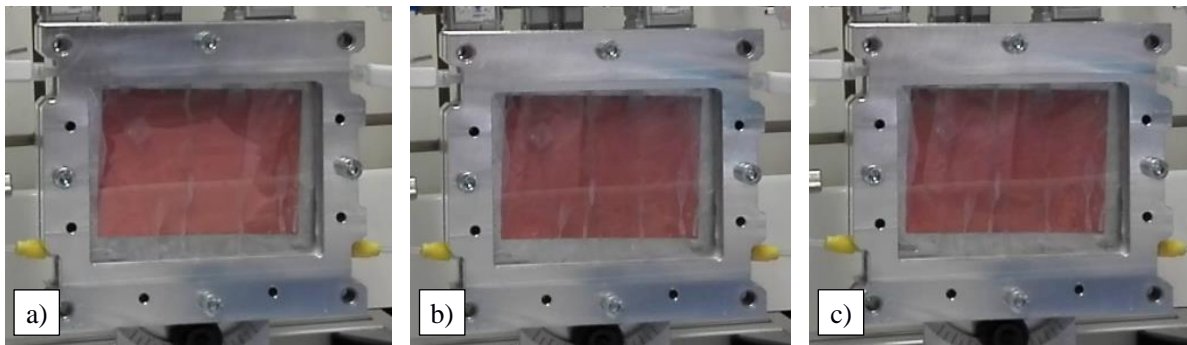


Figure 3: Test 4 – Wetting after a) 2s; b) 5s; c) 10s

## 2.3 Results and conclusion of the preliminary investigation

Considering the production process from ecological and economic aspects, high potential for optimisation was detected at the point of electrolyte filling. Due to the strict air-conditioning requirements when using electrolytes, large dry rooms are necessary to ensure that production is safe. Furthermore, a low dew point of at least  $-70\text{ }^{\circ}\text{C}$  is a precondition for high-quality battery cells. Based on these facts, the dry room should be avoided during filling and all downstream processes. To reduce the negative environmental influence, mini-environments should be used instead of drying rooms.

To guarantee a wide range of applications, the demonstrator should have a modular structure. This is intended to enable a quick changeover and to adapt the device to several cell designs. Furthermore, the demonstrator should provide various adjustment options to find the ideal process parameters for every kind of cell. The experiment evaluation indicates that up to four injection positions should be integrated in the cell and in the device. To ensure that the filling is integrated in a continuous process, a new connection to the cell to inject the electrolyte needs to be developed.

### 3 Development and functional description of the plant concept

Based on the preliminary investigation, a plant concept was developed consisting of two devices: the assembling device and the filling device. The essential aspect of the technology as a whole is the connection between the devices and the cell. This is realized in the form of the newly developed polypropylene port (PP port) (see Figure 4 b)).

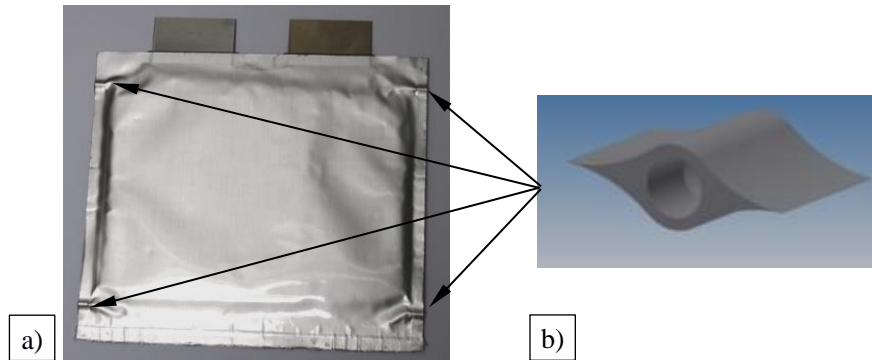


Figure 4: a) Sealed battery cell with ports; b) port

With the aid of this, it is possible to establish a tight connection between the injection needle and the cell. The port is sealed into the seam of the battery cell as shown in Figure 4 a). As the port consists of the same material as the inner layer of the cell film, the connection can be firmly bonded. The surfaces of the film and the port are bonded using a heat-sealing process.

#### 3.1 Assembling device

In the first step of the production process, the battery cell is assembled. This takes place at the first unit, the assembling device (see Figure 5). Here, the individual parts of the cell, such as the port, the electrode stack with current collectors and the film, are correctly positioned then sealed using a heat-sealing process.



Figure 5: Sealing device without enclosure

The sealing process starts with the sealing frames moving towards each other. The silicone-coated, permanently heated sealing jaws are pressed against the film pneumatically. To enable the electrolyte to be guided into the cell, four polypropylene ports are sealed into the seam, into which injection needles can

then be inserted. As a result, the cell is tightly sealed. The three parameters of sealing pressure, temperature and time can be regulated by the user, infinitely variably, before every assembly. This enables an adaptation to different materials (film, ports, current collectors), which guarantees high flexibility. The sealing frame is of a modular design, so the sealing jaws can be replaced. If the cell design changes in the form of a modification of the ports or different cell dimensions, for example, the sealing frame can be modified very easily. At the experimental stage and when adjusting the parameters, different sealing jaw coatings were investigated. Beside the uncoated jaw, coatings with fabric-reinforced Teflon tape and fabric-reinforced silicone tape were tested. Due to its good thermal conductivity, the untreated surface of the sealing jaw quickly heat-sealed the film, the port and the current collectors. However, the disadvantage is that the top polyamide layer of the film sticks to the sealing jaw. The seam was also seen to be bonded irregularly. Due to the inaccuracies in the flatness of the sealing jaws, in some areas there was no contact between the upper and lower surface of the film. The same occurred using the fabric-reinforced Teflon tape, despite its good non-stick properties. Much better results could be achieved using the fabric-reinforced silicone tape. The material used is about 0.35 mm thick. Furthermore, the silicone is more elastic than the Teflon, so full-surface contact between the sealing jaw and the layer is possible.

### 3.2 Investigation of the sealing seam

The sealing seams' strength was investigated using various experiments. To gain an overall impression of the quality and to reveal possible weak spots, a pressure test was carried out underwater. It was seen that the cells were at least macroscopically completely sealed, since no ascending air bubbles were recognizable. The cell burst in the area of the current collector only at an internal pressure of approximately 0.8 bars. This weak spot was found in this test in all the cells under investigation. That means that the port is not the weakest point within the sealing seam.

To examine the sealing seam quality, an investigation was carried out using a tensile test. The sealing seam between film and film, in the area of the ports and in the area of the current collectors, was investigated as shown in Figure 6.

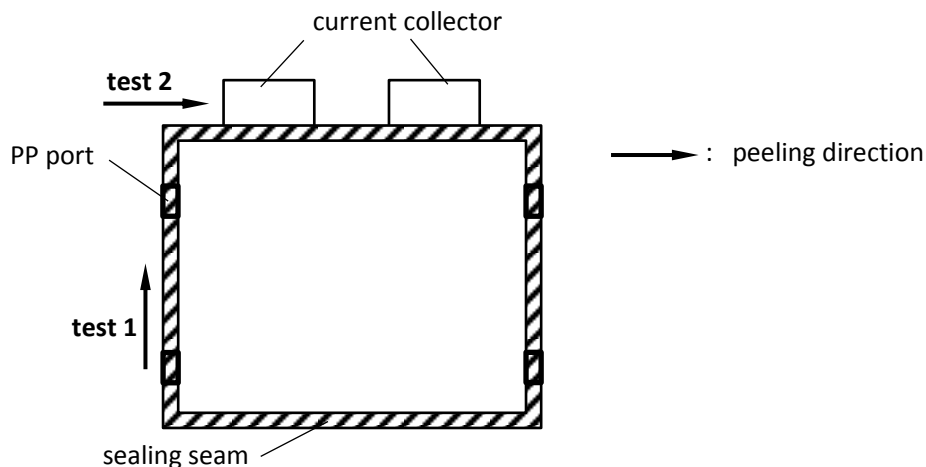


Figure 6: Schematic representation of the sealing seam tests

For this purpose, eight cells were produced with sealing parameters as shown in Table 1.

Table 1: parameter variation – cell sealing

#	1	2	3	4
temperature [°C]	225	230	225	230
sealing time [s]	5	5	7	7

The evaluation of the tests shows that the tensile force follows a nearly parameter-independent course. As an example, an evaluation is carried out for parameter variation #4.

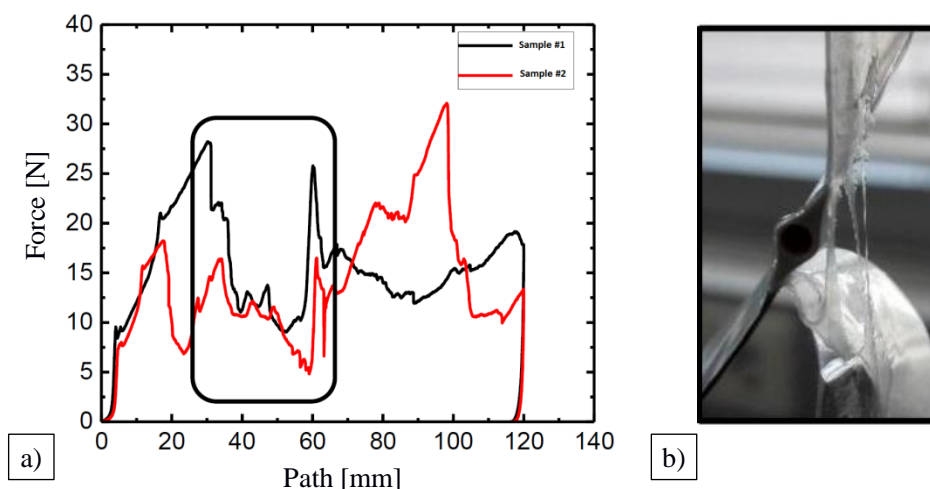


Figure 7: Tensile test 1 – cell sealed with parameter variation #4: a) force-path curve; b) sample #1

Figure 7 shows tensile test 1. Because of the increasing force at the beginning of the test it becomes clear that the film is completely bonded. That can also be seen because the PP coating peels off the film. In the area of the ports a decrease in the force can be seen. This behaviour became evident after 30 mm at the examination of sample #1 (black force-path curve) and after 20 mm at the examination of sample #2 (red force-path curve). Nevertheless, the connection between the port and the film is tight, as proven by the pressure test.

The examination of the sealing of the current collector (see Figure 8) shows almost identical results.

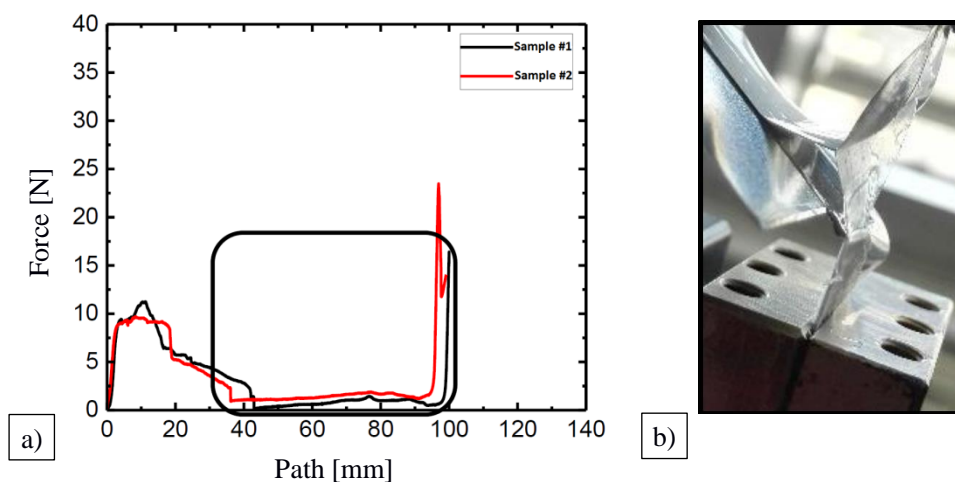


Figure 8: Tensile test 2 – cell sealed with parameter variation #4: a) force-path curve; b) sample #1

At first, an increasing tensile force can be seen. In the areas of the current collectors, the force decreases, meaning that the connection is not as strong as the connection between the films. This suggests that the film and the pp coating of the current collectors are not melted together.

As a third criterion to evaluate the sealing quality, a long-term vacuum test was carried out. For this purpose the cell was evacuated up to a pressure of 150mbar before being sealed. The cell was sealed using the parameters from test #4 (see Table 1). The height of the vacuum in the cell with respect to time-dependent behaviour was assessed optically. Even after 10 months, the film still lies tightly on the test-sealed cardboard. This shows that the sealing seam is sufficiently thick to preserve the vacuum.

### 3.3 Filling device

After their assembly, the prefabricated cells are passed to the second device, the filling device (see Figure 9). A robot grabs the cells from special storage points, which guarantees the utmost in positioning precision. This is enabled by small pins which fit into the holes in the PP port. To make it easier for the needles to penetrate the ports, the ports are drilled at a drilling station up to a remaining wall thickness of about 0.2mm. Subsequently, the cells are transported by the robot to the filling station (see Figure 11), where the ports are penetrated by up to four injection needles. After the cell is evacuated to a pressure of approximately 150mbar (abs) or flooded with different gases to accelerate wetting, it is filled using a dosing pump.



Figure 9: Filling device without enclosure

To implement the findings obtained in the preliminary investigations and depending on the strategy, the filling can take place from different positions to control the electrolyte flow into the cell. At the same time, the pressure can be controlled to affect the wetting behaviour of the electrodes and separators. In addition, the volume flow and the total volume of the electrolyte can be controlled depending on the requirements of the cell.

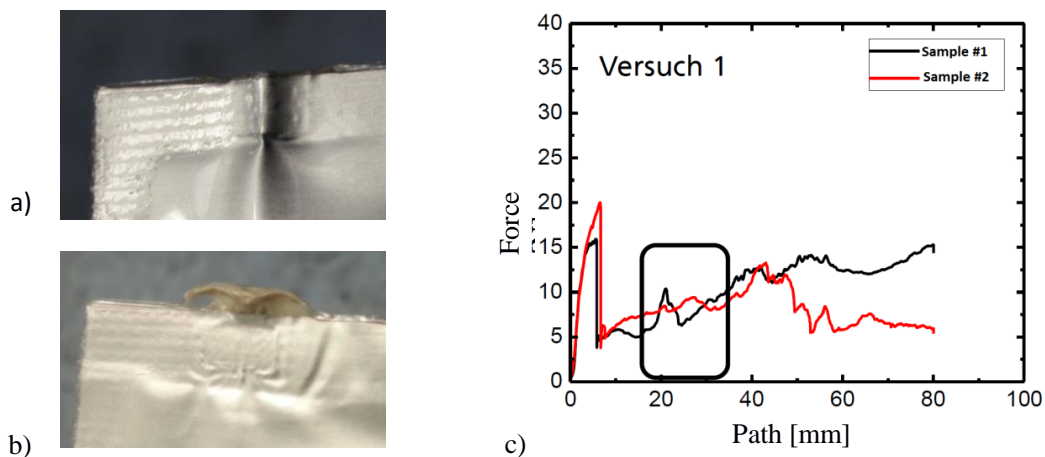


Figure 10: a) pre-sealed port; b) completely sealed port; c) tensile test 1 with completely sealed port

After filling, the pre-sealed and penetrated ports (see Figure 10 a)) are closed by hot sealing. As the superfluous port material is pressed out of the seam by the sealing bars, the seam in the area of the ports is no thicker than the remaining seam (see Figure 10 b)). Now the cell is sealed and prepared for transportation, storage and formation in a standard environment.

### 3.3.1 Investigation of the completely sealed port

To evaluate the quality of the sealing seam at the area of the ports, a tensile test was also carried out. This revealed that there is no decrease in the force in this zone (see Figure 10 c)). That means that the sealing seam is of approximately the same quality as the seam in the area of the films. A further advantage of the PP ports is that there is no foreign material contaminating the seam, because the inner coating of the film is also made of polypropylene.

### 3.3.2 Suction of forming gas

The gas which is a consequence of the formation process can also be sucked out via the injection needles and the ports, making the gas bag superfluous. This process also takes place in the filling station of the filling device (see Figure 11). The same injection needles can be used to suck out the gas as were used to inject the electrolyte. Subsequently, the cell is evacuated by means of the needles and an ejector and sealed at the location of the ports.

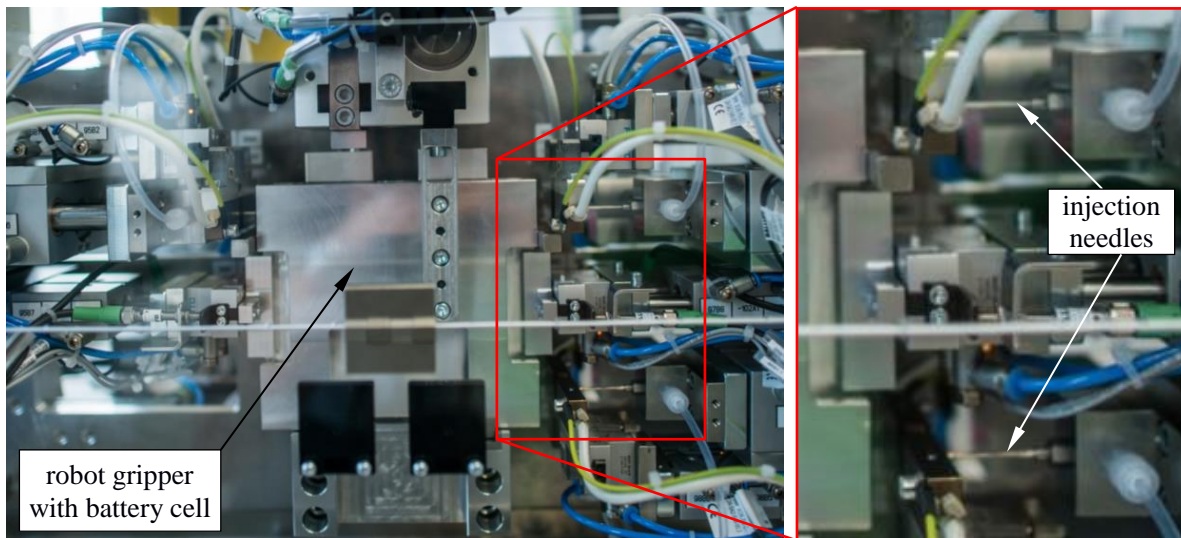


Figure 11: Filling station of the filling device

## 4 Mini-environment

The assembly, the sealing process and also the filling process with subsequent forming gas suction take place within a mini-environment. For process engineering reasons, however, dry air is no longer required for filling and gas extraction because the cells are completely sealed. The enclosure of the system is only necessary for operator safety reasons. Operating materials and products can be fed into and removed from the process by means of a sluice. The air treatment plant achieves dew points of at least  $-70\text{ }^{\circ}\text{C}$ , thereby achieving top production quality. Due to the mini-environment it is possible to reduce the operation costs for the air conditioning, as no dry room is required.

## 5 Conclusion

With respect to the sealing device, several conclusions can be drawn from the experiments and their results. On examining the ports with the tensile test it became clear that there is a need for a targeted heat input to guarantee optimal sealing, as the sealing time depends on the thickness of the materials or their thermal mass. That means that the seam in the area of the PP ports and the current collectors needs more heat than the sealing seam between the films. In summary, it can be noted that the demonstrator is able to produce sealed cells, confirming that the technology functions.

In relation to the filling device, it was confirmed that it is possible to produce sealed cells with the option to control the electrolyte stream and the pressure before, during and after filling. The parameters can be

adapted to a wide range of different cell technologies and dimensions and to the wetting of the active materials in the cell can be controlled. In addition it is possible to extract the forming gas without the need for a gas bag.

Table 1 summarizes the various applied and newly developed technologies and their economical and ecological benefits.

Table 1: Applied technologies and benefits

Applied technology	Economical benefits	Ecological benefits
mini-environment	lower operating costs due to less air conditioning	better environmental balance due to lower energy consumption
port technology – tightly sealed cell after assembly	lower costs for delivery and storage as no need for air conditioning	better environmental balance due to lower energy consumption
port technology – vacuum before filling	lower operating cost due to faster wetting of electrodes and separators	
port technology – suction of forming gas	lower costs for multilayer film as gas bag no longer required	reduced negative environmental impact as less production waste

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