

10th CIRP Conference on Assembly Technology and Systems (CIRP CATS 2024)

Approach to evaluate handling processes of polyethylene oxide (PEO)-based composite cathodes

Ann-Kathrin Wurba^{a,*}, Cedric Bilger^a, Patrick Rehm^a, Jürgen Fleischer^a^a Institute of Production Science (wbk), Karlsruhe Institute of Technology (KIT), Kaiserstr. 12, 76131 Karlsruhe, Germany* Corresponding author. Tel.: +49 1523 9502617; fax: +49 721 608 45005. E-mail address: ann-kathrin.wurba@kit.edu

Abstract

The development of battery technology has made great advances in recent years. However, the increasing demand for electric mobility also highlights the limitations of conventional lithium-ion battery (LIB) technology, such as limited range and fast charging capability, as well as challenges related to battery safety. Among all the technology alternatives, the solid-state battery technology shows great potential to enable higher safety due to the use of non-flammable solid electrolytes instead of liquid electrolytes as well as higher energy densities by replacing the graphite anode with a lithium metal anode. In addition to the trend towards improved battery cell performance, there is also a growing need for the production of application-specific cell geometries. At the Institute of Production Science (wbk), the approach of agile cell production for conventional lithium-ion batteries is currently being investigated. In order to extend the scope of this material and format flexible approach, manufacturing processes for solid-state batteries (SSB) are being considered and further investigated. As a first objective, this work presents a concept for the evaluation of the handling processes of polyethylene oxide (PEO)-based composite cathodes with a special focus on the pick-and-place process. PEO-based composite cathodes are mechanically sensitive, with adhesive and limp characteristics. Therefore, the design of a semi-automated test rig is presented, which allows reproducible gripping, moving and placing of PEO-based composite cathode single sheets and thus stacking on pre-defined platforms. A camera system is integrated to monitor and quantify the process quality. An image processing tool is developed to determine the positioning accuracy and to detect possible damages on the electrode surface from handling. Furthermore, a concept for performing the handling tests is presented.

© 2024 The Authors. Published by Elsevier B.V.

This is an open access article under the CC BY-NC-ND license (<https://creativecommons.org/licenses/by-nc-nd/4.0>)

Peer-review under responsibility of the scientific committee of the 10th CIRP Conference on Assembly Technology and Systems

Keywords: Handling, Monitoring, Solid-state battery

1. Introduction

The worldwide production of solid-state batteries (SSB) is projected to increase from below 2 GWh at present, to 55 - 120 GWh by 2035 [1]. Compared to conventional lithium-ion batteries (LIB), SSB offer superior safety, as the primary fire risk in LIB has its origin in the liquid electrolyte, which is in a solid state in SSB. In addition, the materials used in SSB allow for greater energy density [2]. Moreover, SSB enable new battery designs, which allow the utilization of previously unused space between the individual cells [3]. Despite these promising properties, SSB are still in development on a

laboratory scale. The materials are not available in large quantities and various challenges regarding the electrochemical properties still exist. The most important groups of materials are currently oxide, sulfide and polymer electrolytes, with the group of polymer electrolytes currently being the most developed one and available in the largest quantities [1]. A more detailed comparison between LIB and SSB as well as schematic illustrations are given in [1].

A survey among experts predicts, that between 20 – 60 % of the manufacturing infrastructure from LIB production could be reused for SSB production. The most important factor is the selected material for the SSB, as each material pairing requires

unique production specifications. For example, the production of cathode composites for oxide and sulfide-based SSBs requires a wet chemical process, whereas polymer-based SSBs use extrusion. SSBs also behave mechanically differently to LIBs. Oxide-based solid electrolytes (SE) are brittle, while sulfide-based SE are somewhat softer and more flexible. [1] In contrast, polymer-based SE are highly flexible and show adhesive properties [4]. The group of polymer-based SSB is expected to be the easiest to scale up, while sulfide- and oxide-based SSB will require different machines and process environments. [5] Another issue is the cost producing SSB, as it is currently much higher than that of LIB due to their well-established manufacturing processes. In order to transition to SSB, these costs must become comparable to the current costs of LIB, which is approximately 101 \$/kWh in 2021. [6,7]

These challenging technological uncertainties can be addressed by agile production systems, which combine the flexibility of flexible production systems with the productivity of rigid transfer lines. This is achieved by creating modular functional units that can be added to the production line, as market demand requires. These functional units are able to handle different materials and cell formats.[8] A demonstrator for such a functional unit is shown in [9]. It consists of a microenvironment containing a central handling robot as well as three production modules for stacking, contacting and sealing processes in LIB production. Currently, the demonstrator is only capable of manufacturing LIB pouch cells. [9]

The aim is to test at an early stage and with little material quantities how the materials for SSB can be processed on systems for conventional LIB. Therefore, this paper presents a concept for a semi-automated handling test rig. This will allow an initial assessment of whether the stacking module of the demonstrator mentioned above could in principle also be used for polyethylene oxide (PEO)-based composite cathodes.

The precision of single sheet stacking significantly affects the battery performance as stated in [10], where a direct correlation has been found between the electrochemical performance of a battery cell and the overlap of the individual sheets during stacking. Consequently, accuracy is a vital metric for evaluating a handling system. Hence, the test rig is specifically designed to assess the accuracy of sheet placement, while a critical consideration for PEO-based composite electrodes due to their adhesive nature is necessary. This adhesive property poses challenges in the gripping process, leading to difficulties in detachment. Therefore, special methods such as release mechanisms are envisioned to address this issue. Furthermore, it provides the evaluation of the single sheets surfaces for possible damages caused by handling using image processing.

A test rig similar to the one described in this work, which investigates the gripping behaviour of SSB, is shown in [11]. The kinematic system with a fixture for various grippers is housed within a glove box, that allows the use of inert gas. The electrodes are placed on a turntable while a camera is mounted above the turntable. For quality control, an image processing tool calculates the position and orientation of the electrodes. [11] Using the previously introduced test rig of [11] five distinct grippers for PEO-based cathodes are compared in [4].

Two vacuum suction grippers with varying contact surfaces, a Bernoulli gripper and two electrostatic grippers featuring different dielectrics were evaluated. The grippers were then tested with different moving velocities, gripping distances and settings regarding the pose and orientation repeatability. The electrostatic gripper with PTFE as the dielectric showed the most favourable results. [4]

In the following, the concept for the handling test rig developed in this work is presented in section 2 while the concept for the image processing tool is shown in section 3. Section 4 gives an overview of an exemplary test procedure.

2. Concept for the Handling Test Rig

In this section the concept for the handling test rig is shown. The concept was developed according to the VDI 2221 [12] guideline, starting with a requirements analysis. Afterwards the operating principles and the derivation of the modules are presented. Finally, an overview of the system control architecture is provided.

2.1. Requirements Analysis

The main goal of this work is to establish a testing platform for evaluating the stacking process in the solid-state battery cell assembly with the focus on handling of PEO-based cathodes. The purpose of the test rig is to provide a consistent and repeatable process in which the PEO-based cathode single sheets can be gripped, moved and placed by a gripper. Specifically, these sheets are arranged as a stack, with the gripper gripping only the top sheet. Special attention is given to preventing the formation of wrinkles or creases when gripping individual sheets. Additionally, a design restriction for the gripper is, that it must replicate the shape found in reference [8] for a thorough examination of its usability. One of the main challenges and key differences from stacking electrodes for conventional LIB is that the PEO-based cathode surface has adhesive properties. The experiments should be performed in dry room conditions. Additionally, an automated stacking process while monitoring the orientation, position and defect formation of the sheets is to be ensured. Moreover, a modular structure is essential to provide the flexibility required when changing the geometry of the individual sheets. All of these constraints must be carefully considered throughout the entire system design process, as it must adhere to predefined space constraints, while prioritizing ease of use and efficient operation.

2.2. Operating Principles

Before the stacking process begins, the PEO-based cathode stack needs to be manually placed on the receiving platform. Then, the top single sheet is gripped by the gripping unit and placed on the stacking platform. This seamless operation is facilitated by the continuous tracking of the gripper's position throughout the process. The clamping module is responsible for releasing and securing the sheets on both the receiving and stacking platforms. Subsequently, the gripping system returns to its initial position to restart the process and to guarantee its

repeatability, as mentioned in 2.1. The process is completed when the predetermined number of single sheets has been stacked on the stacking platform. Throughout the entire process, a camera system monitors the position and orientation of the sheets, as well as surface defects caused by handling. For a more in-depth description of the test procedure, a detailed flowchart is depicted in section 4.

2.3. Subdivision of the Modules

As indicated in the design restrictions outlined in section 2.1 implementing a modular design for the test rig is essential to enable rapid adaptation to different scenarios, including changes in sheet size and electrode material. This modularity covers the transport module, the gripping module, the quality control module and the clamping module, all shown in Fig. 1.

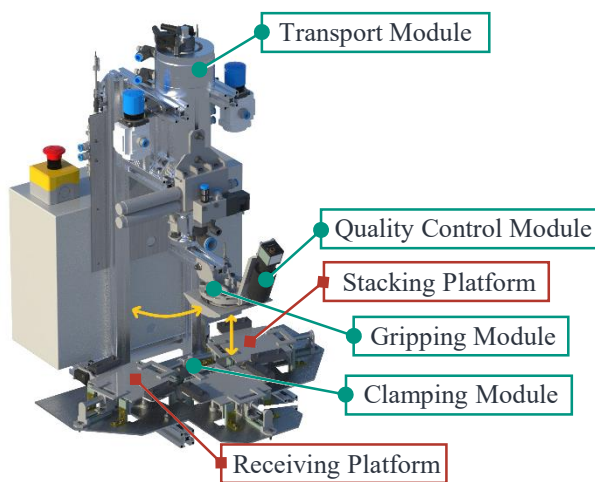


Fig. 1 Overview of the subdivision of the modules of the test rig, including the transport module, gripping module, quality control module and clamping module

According to the requirements outlined in section 2.1, the transport module is dedicated to achieving precise and repeatable movements. Therefore, a commercially available pneumatic linear-rotary unit was installed to perform both translations and rotations, shown as yellow arrows in Fig. 1.

The gripping module is responsible for securely and accurately handling individual sheets during the stacking process. Hence a vacuum-based area gripper is designed for gripping and placing the sheets. Most of its components are 3D-printed, making it adaptable to various sheet geometries. Additionally, the choice of a curved shape is inspired by the gripper of the demonstrator presented in [9], whose suitability is to be investigated. The idea behind employing the curved shape is to effectively reduce the risk of wrinkling in the individual sheets, as highlighted in the design restrictions in section 2.1. This design choice enables the sheets to roll up more smoothly on the gripping surface.

Fig. 2a) shows an exploded view of the gripper and b) illustrates the gripping process using the curved gripper as described. In order to overcome the adhesive properties of the PEO-based cathode sheets, mentioned in section 2.1, a non-stick PTFE film is attached to the gripping surface.

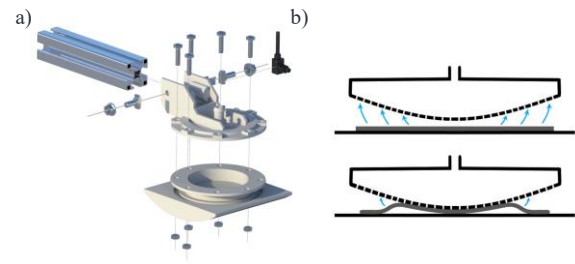


Fig. 2 a) Exploded view of the gripping module, including the gripper arm. b) demonstration of the gripping process for an individual sheet using the curved area suction gripper to prevent creasing.

The clamping module is responsible for securing the stacked sheets after they are placed on the stacking platform and releasing them when they need to be picked up at the receiving platform. This functionality is essential to guarantee that the position and orientation remain consistent, as emphasized in the requirements outlined in section 2.1. This critical function is achieved through a purely mechanical approach. When the gripper applies pressure onto the platform, the direction of this force is transformed by smoothly sliding on inclined surfaces positioned between the platform and the clamping module. As a result, the clamp, which is responsible for holding each sheet in place, loses contact with the stack and moves aside, allowing the gripper to pick up the next sheet in line. As the gripper ascends again, the entire process reverses itself in a seamless manner. One of the clamping modules and the associated platform is shown in Fig. 3.

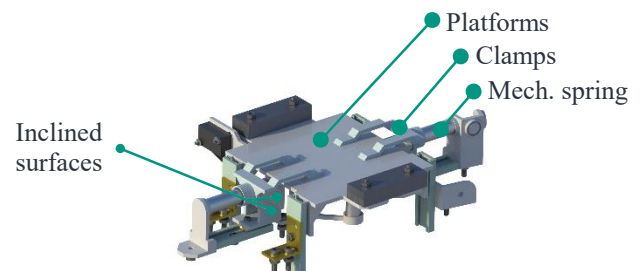


Fig. 3 Overview of the integrated clamping module

Finally, the quality control module is responsible for monitoring and maintaining an overview of both the position and orientation of the sheets throughout the stacking process as well as for the detection of defects from handling. This is achieved through a camera system that provides a live image feed, allowing for the analysis of the sheet location via image processing techniques. Details are presented in section 3.

2.4. System Control

The system control is carried out by a microcontroller. It receives and processes positional data from the gripping module to ensure precise movement of the transport module. Furthermore, the microcontroller's role extends to managing vacuum supply for the gripper, deciding when to activate or deactivate it. This comprehensive control is made possible by the microcontroller controlling both the pneumatic components in the transport system and the gripping module, actuated by magnetic valves. These magnetic valves are operated via a transistor circuit, regulating both the motion of the transport module as well as the vacuum depending on the grippers

position. This positional data is acquired through sensors, specifically, tactile sensors that detect the gripper's alignment using various reference points such as the platforms, its highest position and the required rotation. As mentioned above, the clamping module operates in isolation from the microcontroller, relying solely on mechanical mechanisms for its functionality.

2.5. Initial Testing

Initial tests during the commissioning and optimization phase showed that it is possible to pick up and place the cathode sheets without wrinkling as presented in Fig. 4a). The white streaks on the back of the sheet have their origin in the manufacturing process and have nothing to do with the gripping tests. Furthermore in Fig. 4a), it is displayed, that the cathode fitted very well to the gripper. The PTFE film applied to the gripper surface, has been punctured at the specific points aligning with the vacuum holes in the gripper. This caused slight unevenness on the gripper surface, which can also be recognized on the back side of the sheet when gripping. Slight indentations on the coating are also noticeable, as shown in Fig. 4b). The application of the PTFE film needs to be optimized in future work to prevent these indentations.

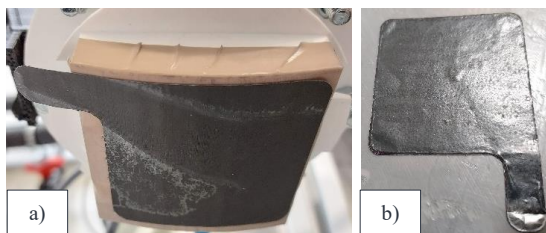


Fig. 4 a) Back side of the gripped cathode sheet, b) coating surface after gripping

3. Concept for Monitoring the Positioning Accuracy and Damage Detection

The primary objective of the monitoring system is to provide highly accurate information of the positioning and surface damages of the PEO-based cathode single sheets. Additionally, the system needs to be able to monitor all the stacking platforms in respect of the spatial constraints.

Considering these requirements, the monitoring system is mounted directly on the gripper. This has the advantage that the monitoring system observes the stacking platform on which the gripper is currently located. In addition, the proximity of the monitoring system to the stacking platform increases its accuracy. However, a disadvantage is the inability to observe all stacking platforms simultaneously.

3.1. Equipment

The camera system components were chosen according to the field of view to ensure full coverage of the stacking platform. The camera sensor selected is the U3-3890CP Rev.2.2 (IDS Imaging Development Systems GmbH), featuring a resolution of 12 MP. It is controlled via a single USB 3.0 connection. The chosen lens is the IDS-10M11-

C1220. This lens has a low image distortion of only 0.77 %. Python was selected as programming language, while OpenCV is used as the primary image processing library in this monitoring system. After image processing, the image resolution is 6.82 MP and the measuring resolution is $0.04747 \text{ mm} \cdot \text{Px}^{-1}$.

3.2. Image Processing Pipeline

In Fig. 5 the proposed pipeline for image processing is presented.

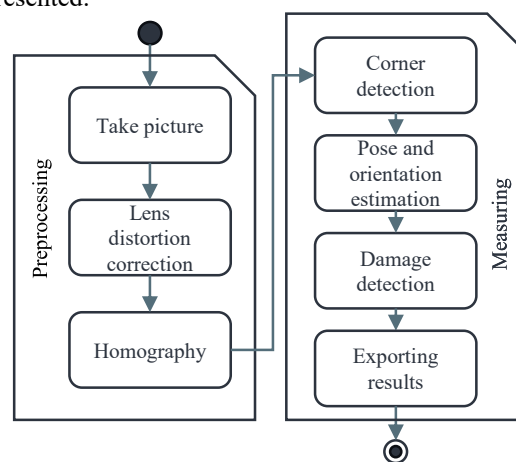


Fig. 5 Flow chart of the image processing pipeline

As shown in the flow chart in Fig. 5, the pipeline can be split into two main stages. Firstly, a preprocessing stage is used to remove distortions from the camera lens and adjustments are made to correct the camera's perspective. Then the measurement steps are applied to the image.

The first step of the preprocessing is the correction of the lens distortion. The distortion coefficients are estimated in advance through camera calibration. The camera calibration was performed using a 7×4 chessboard pattern with 30 different poses. Once the correction of lens distortion is complete, a homography is estimated to obtain a bird's-eye view of the stacking platform. Without a change in perspective, parallel lines in the physical world appear non-parallel in the image. The homography is solved using the direct linear transformation method with four marker points. These circular markers are placed on the clamps of the clamping module of each stacking platform which is shown in Fig. 6. The radii of the circular markers vary between the stacking platforms to distinguish them on the image. The original image and the transformed version can be seen in Fig. 6 a) and b).

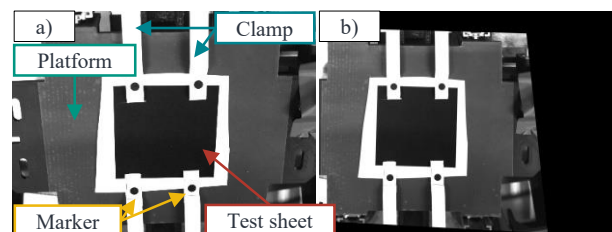


Fig. 6 Comparison of the image a) before the homography step and b) after the homography step

As a first step of the measurement, the corners of the single sheet are determined. For the PEO-based cathode sheets with

an area of 50 mm x 50 mm, a median filter is initially applied to smooth the image. Then, a Canny edge detection algorithm is performed. Using the detected edge points, a Hough transform is performed to approximate a linear function of the edges. Afterwards, the straight lines are classified as either horizontal or vertical lines through k-means clustering. This step is crucial, because the output of the Hough transform may not provide completely parallel lines. The straight lines are visualized as red lines in Fig. 7. Subsequently, the intersections of the horizontal and vertical lines are calculated and also drawn into the image, shown as green crosses in Fig. 7.

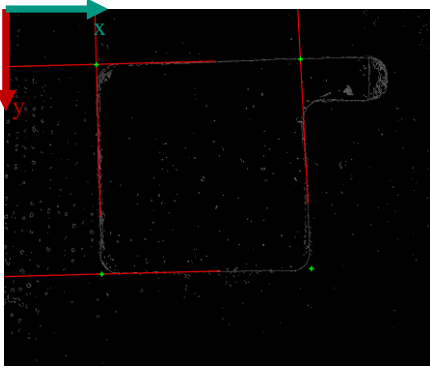


Fig. 7 Image with the result of the Canny edge detection algorithm (white lines), the detected lines from the Hough transform (red lines) and the detected intersections (green markers). The coordinate system is located in the upper left corner.

From those corners, the centre point of the cathode C_c is calculated by averaging the x- and y-coordinates of the corners. The formula for the x-coordinate of the centre of the cathode x_c is shown in equation (1). The calculation of the y-coordinate y_c is done accordingly.

$$x_c = \sum x_i / 4 \quad (1)$$

As a reference, the centre point of the corresponding stacking platform C_p is also calculated analogously to equation (1) by averaging the x- and y-coordinates of the marker points on the clamps. The Euclidean distance d_e between the cathode centre C_c and the platform centre C_p is calculated using equation (2).

$$d_e = \sqrt{\Delta x^2 + \Delta y^2} = \sqrt{(x_c - x_p)^2 + (y_c - y_p)^2} \quad (2)$$

As shown in Fig. 8, the orientation deviation is determined using the top section of the image.

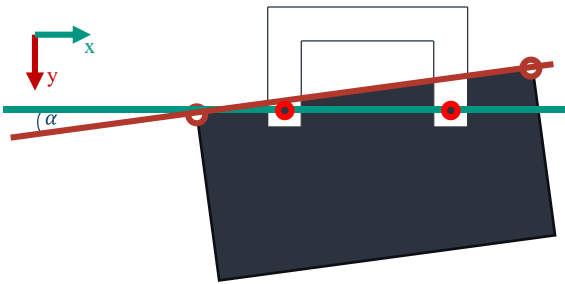


Fig. 8 Visualization of the calculation of the orientation deviation

The slopes m_c of a linear function for the top two corners of the cathode and the two marker points on the clamps m_p are calculated using (3) to determine the cutting angle α which defines the orientation of the cathode in relation to the stacking platform.

$$\tan^{-1} \left(\frac{m_p - m_c}{1 + m_p \cdot m_c} \right) \quad (3)$$

The functionality of the algorithm was verified by extracting the image coordinates of the detected corner points and the markers using an image processing software. Afterwards α and d_e were recalculated according to (2) and (3). A deviation of 0.001° was found for α and a deviation of 0.02 mm was determined for d_e . The differences are mainly due to the inaccuracy of the manually determined image points. The functionality is therefore assumed to be given.

Damages on the cathode surface occur, when the adhesive surface is removed from the gripper surface. The reflection properties are changed and therefore in the captured image the damages appear as brighter areas on the cathode surface. The detection of other damages such as wrinkles are part of future investigations. For the detection of the damages from adhesion, the cathode is cropped from the image by displaying only the pixels inside the four detected corners. Then, the Otsu thresholding method is used to segment the image into damaged and undamaged areas. The threshold is determined by maximizing the variance of the areas above and below the threshold [13]. The extent of damage is determined by calculating the percentage of the damaged area in relation to the total area. The result of performing the Otsu thresholding method can be seen in Fig. 9b), where damages are displayed in white and undamaged parts are black. Fig. 9a) shows the raw image, on which the damage appears slightly lighter.

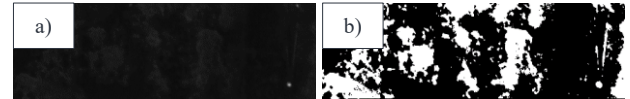


Fig. 9 a) Damaged cathode surface and b) detected damages highlighted in white

In the final stage, the stacking platform number, the Euclidean deviation, the orientation deviation and the percentage of the damaged area are exported to a CSV file for documentation and further evaluations, that are part of future experimental work.

4. Concept for Conducting an Experiment

Fig. 10 illustrates a representative sequence of the stacking process. Initially, the stack is manually placed on the receiving platform and the corresponding number of individual sheets is relayed to the microcontroller. This parameter is adaptable and can be modified to reflect changes in the number of sheets within the stack. Subsequently, the gripper moves to the initial position above the receiving stack. Before gripping the top sheet, an image is captured, which is then further processed. Once the individual sheet has been gripped, the gripper moves to the stacking platform where the stack is formed. After deposition, another image is taken to determine any position and orientation deviations or damages resulting from the handling operation. This process is repeated until the receiving stack is empty. The velocity of the pneumatic actuators can be fine-tuned through a combination of system pressure control and mechanical speed reducers. All experiments with PEO-based cathode single sheets should be performed under dry

room conditions, as mentioned in the requirements list in section 2.1.

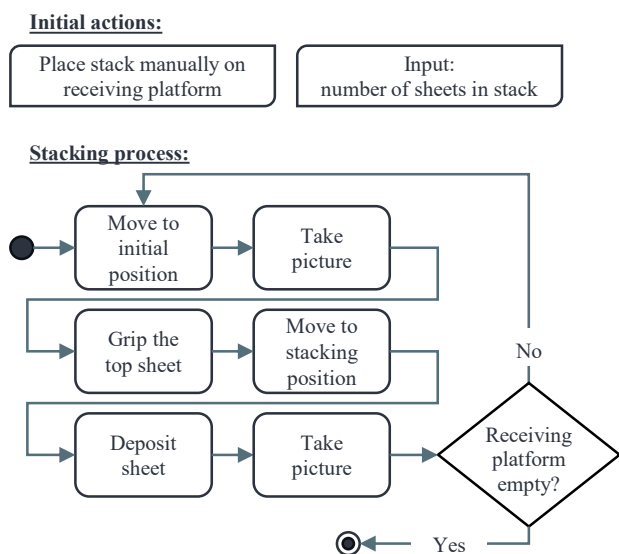


Fig. 10 Visualization of the concept for performing an experiment

5. Conclusion and Outlook

A concept for a semi-automated test rig to investigate the gripping, moving and placing of PEO-based cathode sheets was presented. The test rig is modularly designed with a curved vacuum suction gripper and a camera system with an image processing tool for the position and orientation monitoring and defect detection. The image processing tool is programmed using Python and OpenCV. An exemplary test procedure shows how future experiments will be performed. A pending experimental study will enable the evaluation of the suitability of the demonstrator of [9] regarding stacking PEO-based cathodes. The reliability of the algorithm for recognizing positioning and orientation compared to the reality still needs to be investigated. For this purpose, images could be taken with a 3D scanner (Carl Zeiss GOM Metrology GmbH), which could be used to precisely determine the position and orientation of the sheets. This will enable an evaluative comparison.

Future work will include improving the damage detection to provide clearer results. Furthermore, after validation a series of experiments will be conducted to gain a deeper understanding of the gripping behavior, including the utilization of different gripping heads, the examination of various gripper geometries as well as the operation with different operating pressures and flow velocities. In addition, the test rig could be used and optimized to simultaneously stack the anode sheets of a PEO-based SSB to investigate a scenario closer to industry. Another important focus should be the development of an efficient separation process of the cathode sheets stacked in the magazines before assembly. Future software development for the test rig can incorporate a sophisticated system for automatically recording the sheet number and an automated stopping mechanism that terminates the process when there are

no sheets left on the platform or if the required stack size is reached. Finally, an adaptation of the system to different cell geometries could also be considered.

Acknowledgements

This research was funded by the Federal Ministry of Education and Research (BMBF), Project FB2-Prod (grant number 03XP0432D) within the Cluster of Competence for Solid-state Batteries. This work contributes to the research performed at KIT-BATEC (KIT Battery Technology Center) and at CELEST (Center for Electrochemical Energy Storage Ulm Karlsruhe). The authors would like to thank Laura Helmers (TU Braunschweig) and Frederieke Langer (Fraunhofer IFAM) for manufacturing the cathode samples.

References

- [1] T. Schmaltz, T. Wicke, L. Weymann, P. Voß, C. Neef, A. Thielmann, Solid-State Battery Roadmap 2035+. <https://doi.org/10.24406/PUBLICA-68>.
- [2] L. Helmers, L. Froböse, K. Friedrich, M. Steffens, D. Kern, P. Michalowski, A. Kwade, Sustainable Solvent - Free Production and Resulting Performance of Polymer Electrolyte - Based All - Solid - State Battery Electrodes, *Energy Technol.* 9 (2021) 2000923. <https://doi.org/10.1002/ente.202000923>.
- [3] Y. Kato, S. Hori, T. Saito, K. Suzuki, M. Hirayama, A. Mitsui, M. Yonemura, H. Iba, R. Kanno, High-power all-solid-state batteries using sulfide superionic conductors, *Nat Energy* 1 (2016). <https://doi.org/10.1038/nenergy.2016.30>.
- [4] D.M. Nguyen, T. Scharmann, K. Dröder, Material-adapted Gripping and Handling of PEO-based Cell Components for All-Solid State Battery Cell Stacking, Preprint, TechRxiv (2023). <https://doi.org/10.36227/techrxiv.23696595.v2>.
- [5] F. Duffner, N. Kronmeyer, J. Tübke, J. Leker, M. Winter, R. Schmuch, Post-lithium-ion battery cell production and its compatibility with lithium-ion cell production infrastructure, *Nat Energy* (2021). <https://doi.org/10.1038/s41560-020-00748-8>.
- [6] J. Schnell, T. Günther, T. Knoche, C. Vieider, L. Köhler, A. Just, M. Keller, S. Passerini, G. Reinhart, All-solid-state lithium-ion and lithium metal batteries – paving the way to large-scale production, *Journal of Power Sources* 382 (2018) 160–175. <https://doi.org/10.1016/j.jpowsour.2018.02.062>.
- [7] J.T. Frith, M.J. Lacey, U. Ulissi, A non-academic perspective on the future of lithium-based batteries, *Nature communications* 14 (2023) 420. <https://doi.org/10.1038/s41467-023-35933-2>.
- [8] J. Fleischer, F. Fraider, F. Köbler, D. Mayer, F. Wirth, Agile Production Systems for Electric Mobility (2022). <https://doi.org/10.5445/IR/1000149269>.
- [9] T. Storz, L. Schild, M. Vogt, J. Hofmann, J. Fleischer, G. Lanza, Entwicklung eines flexiblen Produktionssystems für Batteriezellen, *Zeitschrift für wirtschaftlichen Fabrikbetrieb* 116 (2021) 247–253. <https://doi.org/10.1515/zwf-2021-0049>.
- [10] R. Leithoff, A. Fröhlich, K. Dröder, Investigation of the Influence of Deposition Accuracy of Electrodes on the Electrochemical Properties of Lithium - Ion Batteries, *Energy Technol.* 8 (2020) 1900129. <https://doi.org/10.1002/ente.201900129>.
- [11] A. Fröhlich, S. Masuch, K. Dröder, Design of an Automated Assembly Station for Process Development of All-Solid-State Battery Cell Assembly 51–62. https://doi.org/10.1007/978-3-030-74032-0_5.
- [12] VDI 2221 - Entwicklung technischer Produkte und Systeme - Modell der Produktentwicklung, Beuth Verlag GmbH, Berlin 03.100.40, 2019-11-00.
- [13] R.C. Gonzalez, R.E. Woods, Digital image processing (2018) ISBN: 1-292-22304-9.