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Procedia CIRP 130 (2024) 1523–1528

www.elsevier.com/locate/procedia

57th CIRP Conference on Manufacturing Systems 2024 (CMS 2024)

A Novel Machine Concept for the Continuous Manufacturing of 7-layer Membrane Electrode Assemblies for PEM Fuel Cells

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Abstract

The main cost driver for the low temperature polymer electrolyte membrane fuel cell (LT-PEMFC) is the 7-layer membrane electrode assembly (MEA) consisting of one catalyst coated membrane (CCM), two subgaskets and two gas diffusion layers (GDL). To fulfil the auxiliary functions such as gas crossover prevention and electrical insulation, the subgasket area is also referred to as the non-active area, which doesn't contribute to electricity generation. To avoid scraps of cost-intensive materials, the size of the CCM and GDL is often designed only slightly larger than the active area, corresponding to the central cut-outs of the subgasket. Over the last decade, numerous patents have been filed to contribute to the machine and process design to automatically manufacture the 7-layer MEA, which can be broadly divided into pick-and-place based sheet-to-roll processes and traditional roll-to-roll processes, however, with material waste. Pick-and-place operations for fragile and limp MEA components that require damage-free handling are challenging. Hence, a roll-to-roll process for the MEA assembly is still desired given the upcoming manufacturing scale-up. In this work, a novel machine concept based on a cut-and-place principle was proposed and developed, which combines the advantages of efficient material utilization from single-sheet handling and high productivity through adaptive roll-to-roll processing. The design focused on a multifunction-integrated handling system that couples the final manufacturing step with the assembly process to avoid changing the order state of the limp materials during transport and buffer storage. A complete process workflow focusing on the speed synchronization between machine components is also illustrated. Application scenarios and the system limitations of the developed machine concept are also discussed.

Keywords: membrane electrode assembly; cut-and-place; sheet-to-roll process; machine concept design

1. Introduction

During the global transition to zero-emission vehicles, battery-electric vehicles (BEV) were developed to the level of market readiness for the passenger car sector. However, the proton exchange membrane (PEM) fuel cell is still characterized by its high gravimetric energy density and high charging rate, and can therefore contribute more than batteries to the decarbonization of long-distance heavy-duty trucks [1].

The commercialization of fuel cell vehicles is nowadays still in the early stages. One of the most important obstacles is the cost of the fuel cell stack and the resulting lack of market acceptance. The main cost driver for a PEM fuel cell is the membrane electrode assembly (MEA), with almost 80 % share of the total material cost of a fuel cell stack [2]. The current development of production processes in the fuel cell industry is geared towards supplying the market with low volume, which in turn increases manufacturing costs.

Hence, the manufacturing machine for MEA should be designed for medium batches and be scalable without new capital investment. Quality assurance and constituency must be prioritized when designing a machine to reduce scrap costs.

10.1016/j.procir.2024.10.277

The so-called 7-layer MEA consists of one catalyst coated membrane (CCM), two subgaskets and two gas diffusion layers (GDL). Physically, the 7-layer MEA is an assembly out of 5 thin layers. However, to distinguish the function of the proton exchange membrane and the anode/cathode catalyst layers, the CCM is often referred to as a 3-layer MEA. A particular challenge for the automatization of the assembly process lies in the design features. The overall size of the MEA is defined by the size of the subgasket, which includes a central cutout for the active area and a non-active area for the sealing function and gas flow manifolds. Therefore, the size of the CCM and GDL is often designed to be only slightly larger than the active area, so that the excessive material over the active area is just sufficient for the joining purpose. This concentric arrangement of different sizes of rectangular limp sheets has raised challenges for the assembly process. A schematic illustration of the assembly process of 7-layer MEA is shown in Fig. 1.

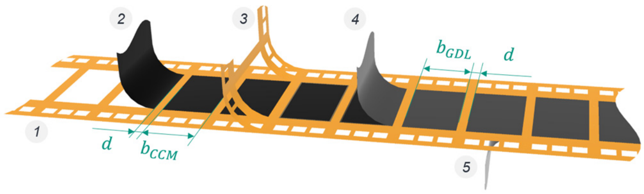

Fig. 1. Schematic illustration of the assembly process of 7-layer MEA (1: lower subgasket, 2: CCM, 3: upper subgasket, 4: GDL-anode, 5: GDL-cathode)

2. State of the Art

2.1. Sheet-to-Sheet

The raw materials of the MEA, namely plastic films, coated membranes and coated felt/paper, are typically produced as roll goods. In the early development phase of fuel cells, the sheet-to-sheet (S2S) approach was often used to manufacture MEA. The roll goods are cut into single sheets with final size and then assembled sheet after sheet into the MEA. Different cutting technologies have been reported, such as flatbed die-cutting [3], rotary die-cutting [4], shear cutting with circular blade [5] and laser cutting exclusively for GDL [6]. Special attention has been paid to workpiece holder when cutting the CCM to avoid wrinkles and bulges [7] and to the cutting edge quality when cutting the GDL [3]. It should be noted that a certain amount of material waste is inevitable in continuous die-cutting processes without an additional web transport system.

For small and medium-scale production, sheets can either be stored in magazines or transported directly to the assembly line via a conveyor system. The assembly process is achieved by repeated pick-and-place operations. Different gripper designs haven been proposed to handle the three components with widely divergent requirements. In [8], an end effector consisting of four vacuum cups that can move relative to each other in the in-plane directions was introduced with the aim of precisely depositing the sheets through on-the-fly adjustment. In the case of magazine storage, a protective film between each layer is inevitable for singulation purpose and to prevent scratches on the functional coating, which complicated a successful and precise gripping even more. Nevertheless, the damage-free handling of the limp sheets with sensitive surfaces and the speed limitations are still challenges for the pick-and-place operations.

2.2. Roll-to-Roll

In addition to the high process speed, the roll-to-roll (R2R) process also stands out for handling the limp web materials. In [9], a classical R2R laminating process was deployed to laminate the CCM with subgaskets, in which all the three material webs are transported simultaneously at a constant web speed and continuously laminated together. However, in this case, the CCM is also sealed up in the non-active area, which is about 40 % of the total area [10]. Considering the cost of the platinum and PFSA membranes, any unnecessary material employment will increase the final product cost of the fuel cell enormously and hinder the development of the technology in the commercial market.

2.3. Adaptive Roll-to-Roll

Efforts have also been made to combine the two processes. In [4,11], an adaptive R2R process was introduced, in which the CCM/GDL is cut directly out of the web and simultaneously transferred to the subgasket by means of a pair of anvil and vacuum-die roll. Although the rest of the material rest can be collected and directly recycled, additional costs are incurred. In [12,13], the problem of material waste is eliminated with the help of carrier film. The CCM/GDL web adhering to the carrier film is separated by kiss-cut and joined to the subgasket web with the predetermined spacing by accelerating and decelerating the carrier film web. However, this intermittent material flow also poses high demands on precise and agile web tension control systems and will limit the maximum process speed. Furthermore, it is also not clear, whether or not a coating defect will occur during the delamination from the carrier film.

3. Analogies and Derivation of Rough Concept

The problem in the assembly process of the 7-layer MEA can be abstracted to a single question: how can a continuous web *A* be separated into discrete parts and placed directly onto web *B* at a predetermined distance without damaging the sensitive and non-rigid material *A*? This isolated question can be reflected in many other industries, where various solutions have been illustrated. The solutions can be summarized into two categories, one is through start-stop mode of the web *A*, and the other is through the velocity difference between the web *A* and *B*.

The first similar issue can be found in the manufacturing of bandages, where the absorbent pads have to be placed on the central area of adhesive woven fabrics or plastics. In [14], a machine was presented, in which the absorbent pads are fed from a continuous web to a punching station, where the pads are punched out and placed directly onto the adhesive fabric, which is fixed by a vacuum roller. The distance between two pads is achieved by the strokes of the stamping tool, where the conveying speed of the pad web has to be adapted to the stroke time. However, this principle requires a certain stiffness of the materials. The bending stiffness of the CCM (near 0) and GDL are not sufficient for such a feed mechanism.

Another approach, particularly suitable for low-cost products such as envelopes with window, is the so-called slip-and-cut applicator. In this approach, one pair of die-anvil rollers feeds and perforates cross-lines on the window materials and another pair of nip-vacuum rollers with higher speed pulls and bursts the web A into discrete patches and places them on the web B [15]. The distance between two patches is achieved by the speed difference. However, this kind of drag and burst principle can create an extensive tension on the material, which is not suitable for the functional requirements of CCM and GDL. In other applications, the cutting process can be performed separately by any type of cross-cutters, under the condition that the leading edge of the web to be cut is already sliding on the vacuum roller [16,17]. The required distance can be achieved by the time interval between sliding and gripping. However, the sliding mode is still not optimal for components such as CCM or GDL with functional coatings, which are sensitive to scratches.

A slip-free transfer application is also mentioned in [18], which demonstrates a concept of zero waste application of trapezoidal ear tabs onto diapers. As the trapezoidal ear tabs are cut directly next to each other to avoid waste, every second ear tab has to be rotated 180° for correct placement due to the trapezoidal pattern. A so-called ear turner with a series of pucks that can be moved radially was introduced. This radial difference between two adjacent pucks provides a clearance to rotate the pucks. However, the distance between two ears after rotation correction is achieved by a pair of vacuum drums with different diameters.

In [19], a flexible and continuous machine concept is proposed for single sheet stacking of the Li-ion battery, where the centerpiece of the concept are the innovative handling elements. The handling elements, designed as vacuum suction grippers, can be rotated to continuously transport the electrode web. A circular blade is integrated into each set of handling elements to cross cut and separate individual electrode sheets from the web. Radial movement of the handling elements allows the machine to be adjusted for different electrode sizes to meet flexibility requirements without tool changes.

Inspired by the mechanism in [18,19], an innovative machine concept has been developed to manufacture and assemble the 7-layer MEA, in which the utilization of the core materials has been maximized by a special kinematic design.

As shown in Fig. 2, the CCM or GDL roll, assuming coming from slitting process and already has the final web width, is unwound and transported to the handling system. The whole handling system is driven in rotation at a constant angular velocity ω, functionalizing similar as a rewinder. The pivoting frame must be positioned as close as possible to the handling system to ensure a straight web edge before it's cut into individual sheets. The handling system consists of various suction gripper segments that can travel radially and operate in two operating radii. With the smaller radius r_1, all the segments can form a closed shape (round or polygonal), that allows the web to be cross-cut continuously without waste by the cutting unit 1. Each gripper segment should grip a single sheet over its entire surface to avoid any kind of wrinkling or bulging of the thin membrane. With the larger radius r_2, the gripper achieves a higher tangential speed at the placement operation. Synchronisation of the tangential speed of the gripper at second operating radius with the conveying speed of the subgasket web ensures a slip-free placement of the discrete CCM/GDL sheet. In addition, synchronisation of the conveying speeds of the two components is often essential to achieve simultaneous bonding of the two components [20]. The adhesive is applied to the subgasket by bonding unit 2 prior to placement. As the bending stiffness of the CCM is near zero, a sequential deactivation mechanism of the vacuum suction in each griper in clockwise direction ensures the precise placement of the trailing edge.

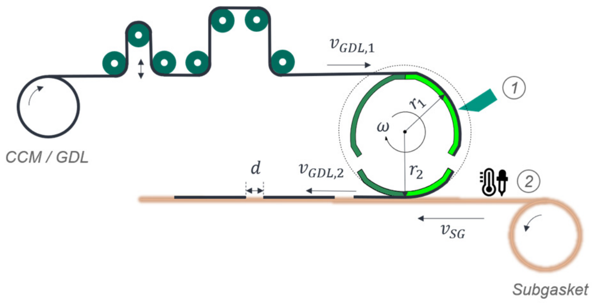

Fig. 2. Schematic illustration of the developed machine concept to manufacture 7-layer MEA without core material scrap

4. Concept Development und Design

4.1. Requirements

A special requirement for the spatial arrangement of the web feed direction of the GDL and subgasket web is related to the GDL intrusion effect. In order to further increase the volumetric energy density and reduce the contact resistance between the layers, the fuel cell stack is compressed to a few megapascal prior to fixation. The compression force causes intrusion of the GDL into the channels, blocking the reactant gas flow and shortening the durability of the cell component.

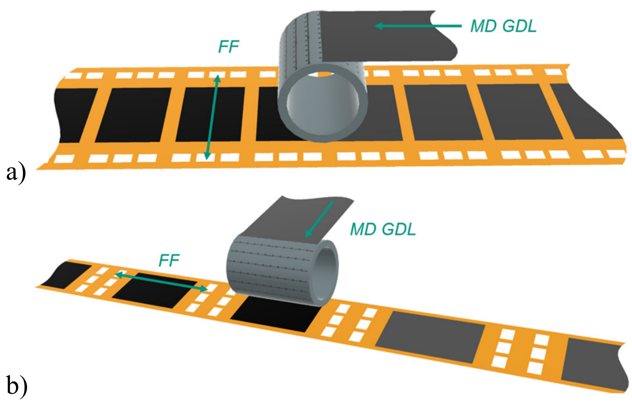

Fig. 3. Scenarios of the spatial arrangement of the GDL and subgasket web (MD: machine direction; FF: flow field direction)

However, the GDL, especially the felt and the paper type, has an anisotropic characteristic in terms of mechanical strength due to the manufacturing process and typically shows a higher stiffness in the machine direction (MD). It has been shown in [21], that by aligning the stiffer direction (MD) of the

GDL perpendicular to the major flow field direction, the GDL intrusion can be minimized, resulting in better performance. Since the major flow field direction of the bipolar plate also defines the orientation of the MEA in a continuous conveyed web by the layouts of the manifolds, there are only two possible spatial arrangements of the GDL and the subgasket web left, as shown in Fig. 3. The scenario shown in Fig. 3.b was not considered further in this work, because the 90° crossing angle of the two coils does not support a synchronized and continuous assembly process.

4.2. Geometrical Design

One way to place a flat object is by designing the gripper into a flat surface to achieve a surface contact. In this case, the multiple grippers together will build up a polygon, such as a triangle, a rectangle, etc., which requires brief pause in the motion and is therefore counter to the objective. Another way to deposit the CCM precisely is to build a rolling mill type movement. Although this only results in line contact, the continuous rolling motion ensures that the entire surface of the CCM can be placed in contact with the subgasket web without any part of it being dropped in an uncontrolled manner.

Since the geometric shape of the gripper should be built as a circular arc, the next question needs to be answered is the curvature radius of the arc R in relation to the two operating radii r_1 and r_2.

If the arc corresponds to the smaller operating radius r_1, then it would be beneficial for the material entry status, as in the pick operation, because the circular shape provides a continuous smooth gripping condition. Conversely, for the placement operation, there is a variable distance b between the real gripper and the fictive circular path (dotted green line) for the placement, as shown in Fig. 4a. In order to compensate for this distance and achieve a line contact between the CCM/GDL and the subgasket, the gripper must move radially synchronized with the angular velocity for less than a few millimeters in few milliseconds, which is very challenging for the control system. Any small delay will cause wrinkles or even cracks in the material.

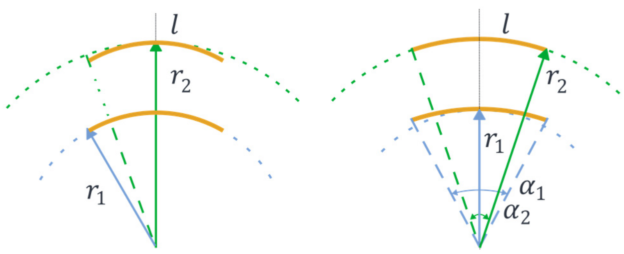

Fig. 4. Geometric design of the gripper: a) R = r_1; b) R = r_2

If the arc corresponds contrariwise to the larger operating radius r_2, the complex adjustment for the placement is eliminated by the circular path, as shown in Fig. 4b. In this case, the material entry faces a similar problem with the changing of the apex of the gripper system, as the grippers are building a form like Reuleaux polygon in the entry track. This variable contact point for the material entry can be compensated by rounding off the edges and state-of-the-art web tension control systems. A simple geometric model was developed to predict the range of the movement for this gripper system. The model is based on the geometry shown in Fig. 4b, where l is the arc length of the gripper and α_2 is the central angle of the arc. Assuming that this handling system consists of n grippers, which ought to form a closed shape in the smaller track, the angle α_1 can be calculated as:

$$n \cdot \alpha_1 = 2\pi \tag{1}$$

Since the gripper must grip a CCM/GDL sheet in its entirety, the arc length l is equal to the width of the final sheet b (shown in Fig. 1). Similarly, the perimeter of the larger track is equal to n times of the sum of the width of the CCM/GDL sheet b (refer to both b_{CCM} and b_{GDL}) and the distance between two sheets d.

$$\alpha_2 = \frac{l}{r_2} = \frac{b}{n \cdot (b + d)/2\pi} \tag{2}$$

The radial travel distance regarding to the central point of the gripper can be found as:

$$\Delta = r_2 \cdot \cos\frac{\alpha_2}{2} - r_2 \cdot \sin\frac{\alpha_2}{2} / \tan\frac{\alpha_1}{2} \tag{3}$$

By varying the radii and angles of the circular arcs, any length and spacing of the CCM sheets can be created. Based on the publicly available data sheets of commercial fuel cell stacks, the active area and total size of the MEA are chosen to be 214 cm^2 and 490 cm^2 for the design of the demonstrator to verify the machine feasibility. More specifically, the width of the CCM/GDL-sheet and the spacing are taken to be 116 mm and 20 mm respectively.

By introducing the MEA size into the geometric design, the maximum deflection of the material entry in the vertical direction can be calculated using Equation (4).

$$\Delta_{r,max} = r_2 \cdot \frac{\sin(\alpha_2/2)}{\sin(\alpha_1/2)} - (r_2 - \Delta) \tag{4}$$

The calculated results of Δ_r and $\Delta_{r,max}$ based on different number of grippers are shown in Tabel 1. If the number of the gripper is too little, not only the time left for the cutting and radial movement won't be sufficient, but also the deflection by the material entry is more significant. If the number of the gripper is larger than 6, there won't be enough installation place for all the machine components. Therefore, 6 gripper system is taken for the demonstrator design.

Tabel 1. Correlation between number of grippers and the deflection of material entry

n	3	4	5	6
Δ [mm]	11.50	14.11	16,99	19.98
$\Delta_{r,max}$ [mm]	4.98	3.55	2.78	2.29

4.3. Conceptional Design of Process Workflow

Assume that the initial state of the whole process is when the central point of the gripper 1 is located at 0° and the whole system rotates clockwise. Two important milestones in the process workflow regarding to one revolution of the gripper 1 is shown in Fig. 5. At the first stage a), the vacuum suction function of gripper 1 is fully activated to grasp the web coming from a lower position compared to the apex point of the gripper. The cutting process can only be initiated, after a whole sheet is "rewind" from the zero point, which corresponds to an angular movement of 30°. Once the cutting process is completed, the gripper 1 can be triggered to travel radially to the second track.

a) Pick: Material entry at 0°	b) Place: Sync begins at 154.4 °

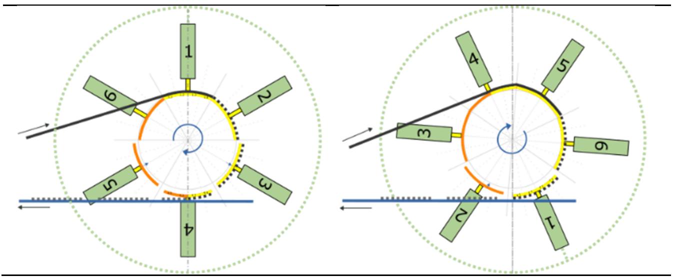

Fig. 5. Process workflow of the novel handling system focusing on one revolution of gripper 1

An essential precondition for the placement operation b) is that the gripper 1 must arrive the larger circular track at latest when the leading edge of gripper 1 reaches the lowest point, which results in an angular movement of the center point of 154.4°. The starting point of the radial movement for the sheet separation can be calculated from the rotation speed ω, the radial travel distance Δ and the speed of the linear axis v_r. If the values are exemplarily taken as 11.5 rpm, 19.98 mm (see Tabel 1), 250 mm/s, respectively, the resulting starting point for the radial movement is 148.9 °. Consequently, the left range for the cutting motion is then between 30 ° and 148.9 °, which must be reached in 1.7 s. It should be noticed that a scale up can easily be achieved by increasing the angular velocity of the handling system. Looking at state-of-the-art servo motor technology, the exemplarity calculation based on 11.7 rpm is way below the nominal speed.

5. Design Implementation and Motion Simulation

5.1. Segmented Suction Gripper

As described in the rough concept in section 3, the rolling placement only results in line contact between the CCM/GDL and the subgasket. Therefore, a successively deactivation of the suction area is desired to avoid uncontrolled falling of the trailing edge. A stepless deactivation of the suction holes is difficult with the radial movement of the grippers. In a normal vacuum roller, the control of the suction area is typically achieved by an additional inner layer with sealing lip, which is not compatible with movable gripper segments. Therefore, a stepwise adjustable vacuum suction mechanism is applied for the current design of the gripper, as shown in Fig. 6. The suction holes are arranged in axial direction and connected to eight rows of long boreholes (shown in sectional view), which are supplied with vacuum or low-pressure air. The low-pressure air supply can then be controlled via valve terminals, where the control command for the suction deactivation simply corresponds with the angular velocity. The number of rows can also be increased under consideration of the installation space limitation for the valves.

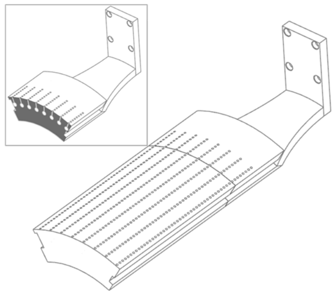

Fig. 6. CAD-model of the gripper with sectional view focusing on internal structure

5.2. Cutting Unit

Based on the rough concept and process workflow, a technology screening is carried out, focusing particularly on the integrability of the cutting technology into the entire machine concept. Laser cutting is not suitable for the purpose of demonstration and validation, because the target focus point for a cross-cut line is changing in all x-, y- and z- directions due to the continuous rotation of the handling system, requiring therefore complicated mirror systems with ultra-precise control. Rotary die-cutting was also ruled out, because the die roller has a high requirement on the concentricity as well as the surface finish of the anvil roller. In the case of this work, the grippers-built reel in non-circular form plays the role as an anvil roller, which is not sufficient for a rotary die-cutting process. Hence, the shear cutting with circular blade was identified as the most compatible cutting technology.

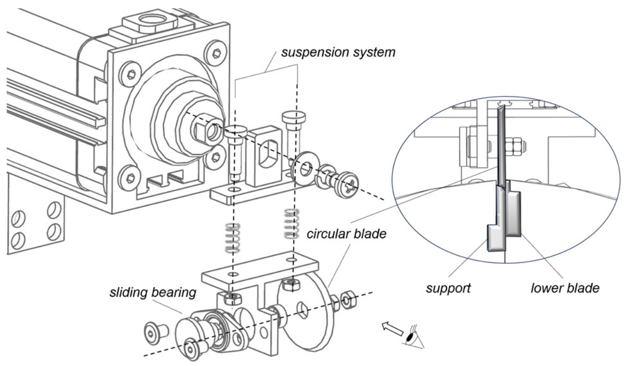

Fig. 7. Partially exploded view of the cutting unit with a front view focusing on circular-lower-blade interface

To achieve a simple linear movement, pneumatic cylinders were selected as linear actuators. Special attention must be paid to collision prevention. This means that the cutting unit should not interfere with either the material entry or the subgasket conveyor during placement operation. The cutting unit is mounted directly on the rotary plate, where the gripper is also mounted on. A telescopic cylinder is chosen to further reduce the installation space need. To build a shear cutting mechanism, a lower blade is implemented as a part of the gripper. Besides the lower long blade, a support piece corresponds to the blade angle is mounted on the adjacent gripper, to guide the circular blade through the whole cutting length and avoid oscillation, as shown in Fig. 7. The circular blade itself is not driven but only rotates passively via a one-sided sliding bearing. A small suspension system is also incorporated into the cutting unit to provide pre-tension to ensure a defined contact between the upper and lower blades.

As described in chapter 2, the workpiece holder is one of the most important factors for a clean cut. In this design, the suction holes beside the upper-lower blade pair hold the workpiece tight. However, the optimum distance between the suction point and the cut line needs to be tested through experiments.

5.3. Motion Simulation

To evaluate the technical feasibility of the designed process, the machine concept is implemented in a 3D model and specified in concrete components. The simulation and validation of the motion between different units is performed using the Mechatronics Concept Designer module of the Siemens NX system. A slow-motion scenario focusing on gripper 1 and its associated cutting unit is simulated and plotted in Fig. 8 (left). The light-yellow area defines the angular range for cutting and radial movement, as calculated in chapter 4.3, which doesn't change with the speed variation. As can be seen from the green line in the plot, the pneumatic cylinder must be driven out to the aim position and back to the home position again, doubling the stroke time, in order to avoid collision with the conveyor for the subgasket. The backward movement of the gripper is relatively relaxing as long as it doesn't interfere with the material entry at 330°. In this scenario, however, the time buffer for the two actions hasn't been fully used. In the end, it is the interaction between the three speeds, i.e., the angular speed, the radial travel speed of the gripper and the cutting speed, that defines the limits of this machine concept.

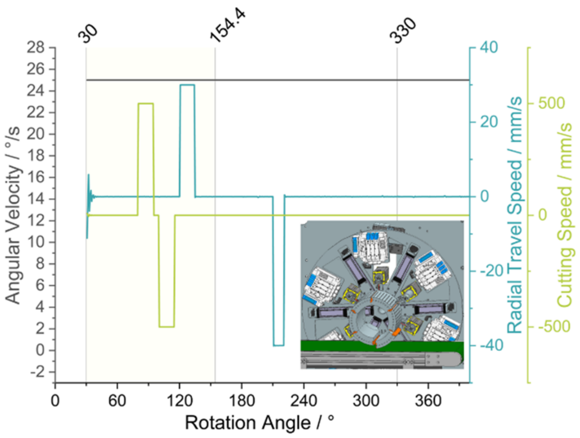

Fig. 8. (left) Plot of operation sequence as a function of rotation angle of the system (right) 3D-Model

6. Conclusion

By reviewing the state-of-the-art of the manufacturing process for 7-layer MEA, a dilemma between optimal material utilization and continuous material feeding and therefore high productivity was identified. By transferring the special design feature, which is responsible for the dilemma, into other industries, partial solutions and inspirations have been found. Consequently, a novel machine concept based on a cut-and-place principle, which combines the advantages of both material utilization and continuous processing has been proposed. A geometric model for the machine concept was developed for the design of motions and workflows. In the end, the essential machine component was designed in detail and implemented in a 3D model. A motion simulation was carried out to validate the feasibility of the process. A scale up can easily be achieved by increasing the angular velocity of the handling system. However, the bottle neck lies on the linear actuator of the cutting unit. In the future work, the demonstrator should be physically built up to test the feasibility of the machine in details, such as the cutting edge quality, placement precision and so on.

Acknowledgements

The authors thank the Ministry of the Environment, Climate Protection and the Energy Sector Baden-Württemberg for the funding of the research project “KontiMEA” under grant no. BWKWHF23107.

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