



# Deliverable Report

Summary Report on material and component manufacturing process

**(D3.12)**

<http://pretzel-electrolyzer.eu/>

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Project 779478 - PRETZEL

Novel modular stack design for high pressure PEM water electrolyzer technology with wide operation range and reduced cost

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## Contents

Contents	4
Abbreviations and Indices	5
1 Summary	6
2 Introduction	7
3 Deliverable in the Project	8
4 Report on Manufacturing Process of PRETZEL stack components	8
4.1 Planar bipolar plates (BPP) with corrosion resistant coating	8
4.2 3D pore-graded porous current distributor (PCD) with macro porous layer (MPL)	11
4.3 Membrane electrode assembly (MEA) based on catalyst coated membrane (CCM) manufacture	12
5 Conclusions	16
6 Appendix	16

## Abbreviations and Indices

Abbreviation	Explanation
BPP	Bipolar Plate
PCD	Porous Current Distributor
MEA	Membrane Electrode Assembly
PEM	Polymer Electrolyte Membrane
MPL	Macro Porous Layer
CCM	Catalyst Coated Membrane
OER	Oxygen Evolution Reaction
PEMEL	Polymer Electrolyte Membrane Electrolyzer
SLM	Selective Laser Melting
CPM	Classical Powder Metallurgy
VPS	Vacuum Plasma Spraying
WP	Work Package
HER	Hydrogen Evolution Reaction
SA	Sigma Aldrich
ARM	ARMINES

## 1 Summary

This public report summarizes confidential descriptions on the entire manufacturing processes of PRETZEL stack components, specifically the bipolar plates (BPP), coated porous current distributors (PCD) and membrane electrode assemblies (MEA). Besides the manufacturing process these reports give attention to problems encountered as well as solutions and the final elaborated results. The development of the inner cell components was aimed at producing components for first test cells with an active area of 4 to 25 cm<sup>2</sup> and the final high-pressure cell design of 500 cm<sup>2</sup> (22.5 x 22.5 cm<sup>2</sup>) active area, which is near industrial scale. Materials needed to be selected, which are able to withstand the respective conditions, especially the highly oxidative environment at the anode side. The objective was to prepare components targeting to a current density of 4 Acm<sup>-2</sup> (6 Acm<sup>-2</sup> peak current density) at potentials below 2 V per cell (2.3 V in overload mode) and under pressurized polymer electrolyte membrane (PEM) electrolysis conditions. For this ambitious purpose the above mentioned cell components needed to be optimized. With the ability to operate at 4 Acm<sup>-2</sup> at rated power and 6 Acm<sup>-2</sup> at overload mode, a production rate of 0.9 m<sup>3</sup>H<sub>2</sub> h<sup>-1</sup> per cell, respectively, 1.4 m<sup>3</sup>H<sub>2</sub> h<sup>-1</sup> per cell can be achieved. It is aimed to develop a short stack with five PEM electrolyzer cells. Due to the homogenous operation conditions for each individual cell in the targeted stack, no relevant upscaling effects are expected. Therefore, the investigated developments give a perspective for megawatt scale PEM electrolyzer systems in the future.

The stack design based on hydraulic single cell compression works with planar monopolar plates. Most important requirements are a high electric and a corresponding high thermal conductivity as well as a certain mechanical flexibility. A high thermal conductivity is required, since the hydraulic fluid is also used for temperature management and, therefore, a good transition of the thermal energy from the process site to the hydraulic medium needs to be guaranteed. The previously mentioned requirements are fulfilled by the developed design, which further guarantees for a long-term stable operation while at the same time contact resistances at the interfaces to the PCDs as well as between neighbouring cells are minimized. The planar structure of the pole plates allows for an easy handling regarding the corrosion protective coatings, which are applied by different techniques.

In contact with the manufactured pole plates are the PRETZEL 3D pore-graded PCD with macro porous layer (MPL). The PCDs were manufactured via two different techniques, on the one hand by selective laser melting (SLM) and on the other hand by classical powder metallurgy (CPM). To prevent oxide formation during heat treatment the necessary sintering was performed in a high vacuum sintering furnace. The produced compound specimens were all over made out of fine Titanium powder for the MPL and coarse expanded metal as well as fiber as support material fulfilling the function to efficiently distribute and remove water and gaseous media to and from the active area, respectively.

The core component of each stack is the MEA, which was prepared by coating of Nafion® commercial membranes with Ir-based anode electrocatalysts and Pt/C cathode electrocatalysts. The coating techniques were applied for delivering electrodes by means of catalyst coated membranes (CCM), i.e. by direct coating on the membrane. The development followed a scale-up concept starting from reference (benchmark) materials and lab-scale techniques towards electrocatalysts developed within PRETZEL project and pilot-scale techniques. Based on processes and results in terms of quality and reproducibility in preparation of electrodes and MEAs as well as their performance, the selected approach for large-scale MEA manufacturing in the project is the preparation of double-CCMs applying the spraying technique.

## 2 Introduction

2018 the worldwide CO<sub>2</sub> concentration was 407.38 ppm while for the pre-industrial time the CO<sub>2</sub> emission was 280 ppm.<sup>1</sup> Only by reducing CO<sub>2</sub> emissions the Paris Accord, which was renewed in 2019 to reinforce 1.5 °C as a safe limit for global warming by the end of this century and net zero emissions by 2050, can be met.<sup>2</sup> In order to achieve this important but ambitious goal, the use of renewable energies is essential and urgently needed. Hydrogen, when produced by water electrolysis, is one of the most promising energy vectors for the large-scale storage of renewable electricity and for decarbonisation of refineries, ammonia production and chemistry industries while linking the energy, industry and mobility sectors.<sup>3,4,5</sup> It can provide a sustainable conversion of electricity to chemical energy to reduce the large share of CO<sub>2</sub> emissions. This places new demands on electrolysis technologies in terms of efficiency, flexibility, performance, long-term stability and costs. Currently, the two most important commercial electrolysis technologies for large-scale hydrogen production are alkaline and polymer electrolytic membrane water electrolysis (PEMWE).<sup>6</sup> Proton exchange membrane (PEM) electrolysis is the most suitable technology given its flexibility, low footprint and wide range of operation. The main disadvantage of PEMWE compared to alkaline systems is the high capital cost which is set more than 30 % higher.<sup>7</sup>

Recent developments in the field of polymer electrolyte membrane electrolyzers (PEMEL) aim on the reduction of overall system costs in order to compete with state of the art alkaline electrolysis. Major optimization aspects refer to an increase of efficiency as well as to obtain high pressure electrolysis. Both issues are related especially to the PEMEL stack, which needs to generate hydrogen with high production rates (high current density) at reasonable cell voltage (efficiency) and high output pressure, so that raw material deployment can be reduced (e.g. noble metals, proton conductive polymers and titanium) and balance of plant components can be omitted (especially, compressor stages). Furthermore, the stack components such as the membrane electrode assemblies (MEA), the porous current distributor (PCD) and the bipolar plates (BPP) need to be mass produced and low-cost designed for an effective use of expensive raw materials. The project “Novel modular stack design for high PREssure PEM water elecTrolyzer tEchnoLoGy with wide operation range and reduced cost – PRETZEL” addresses these aspects to realize the next generation electrolyzer technology meeting the needs of industrial scale hydrogen production in the near future. Beyond these challenges a

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<sup>1</sup> Federal Environment Agency: Atmospheric greenhouse gas concentrations. <https://www.umweltbundesamt.de/daten/klima/atmosphaerische-treibhausgas-konzentrationen#kohlendioxid->, Accessed April 2020.

<sup>2</sup> Climate Action Summit 2019, Report of the secretary-general on the 2019 climate action summit and the way forward in 2020. *United Nations*, 2019.

<sup>3</sup> Appum, K., Sector coupling – shaping an integrated renewable energy system. <https://www.cleanenergywire.org/factsheets/sector-coupling-shaping-integrated-renewable-power-system>, Accessed April 2020.

<sup>4</sup> Adolf, J., Balzer, Ch. H., Louis, J., Schabla, U., Fishedick, M., Arnold, K., Pastowski, A. & Schüwer, D. Shell Hydrogen Study – Energy of the Future – Sustainable Mobility through Fuel Cells and H<sub>2</sub>, *Shell Deutschland Oil GmbH*, (2017).

<sup>5</sup> Robinius, M., Otto, A., Heuser, P., Welder, L., Syranidis, K., Ryberg, D. S., Grube, T., Markewitz, P., Peters, R. & Stolten, D. Linking the Power and Transport Sectors – Part I: The Principle of Sector Coupling. *Energies*, **10** (7), 956 (2017).

<sup>6</sup> Smolinka, T., IndWEDe: Brief Overview – Industrialisation of water electrolysis in Germany: Opportunities and challenges for sustainable hydrogen for transport, electricity and heat. *Federal Ministry of Transport and Digital Infrastructure (BMVI)*, Berlin, 2018, [https://www.now-gmbh.de/content/service/3-publikationen/1-nip-wasserstoff-und-brennstoffzellentechnologie/181204\\_bro\\_a4\\_indwede-studie\\_kurzfassung\\_en\\_v03.pdf](https://www.now-gmbh.de/content/service/3-publikationen/1-nip-wasserstoff-und-brennstoffzellentechnologie/181204_bro_a4_indwede-studie_kurzfassung_en_v03.pdf).

<sup>7</sup> FCH-JU, Addendum to the Multi-Annual Work Plan 2014-2020, *Fuel Cells and Hydrogen 2 Joint Undertaking* (2018).

significant increase of lifetime and improved operability to cope with the intermittent electricity supply from renewable energy sources are also provided.

The overall goal of PRETZEL is to develop an innovative PEMEL that provides significant increases in efficiency and operability satisfying the market demands. Such electrolyzers are urgently needed in the context of the increased demands of the grid balancing market. PRETZEL is offering a break-through in becoming “Game Changer” in the field of water electrolyzers. To reach this overall goal PRETZEL has the following objectives:

1. Develop and manufacture the components of the PRETZEL stack for the innovative high pressure PEMEL that operates at increased temperatures.
2. Develop and manufacture the PRETZEL high pressure PEMEL stack based on the novel principle of hydraulic compression.
3. Set-up and undertake continuous procedures to evaluate the development process through all phases against PRETZEL specifications.
4. Integrate the innovative PEMEL stack into a high pressure PEMEL test facility and validate the overall performance and operational criteria.
5. Disseminate and exploit the innovations in PRETZEL in order to prepare the market penetration of the new technology.

### **3 Deliverable in the Project**

Manufacture of inner cell components for initial test cells with an active area from 25 to 100 cm<sup>2</sup> and the final high-pressure cell design with an active area of ca. 500 cm<sup>2</sup> (22.5 x 22.5 cm<sup>2</sup>). The component design needs to be feasible for the targeted peak current density of 6 Acm<sup>-2</sup>. The PRETZEL consortium aims to develop the following components:

- Planar BPPs with corrosion resistant coating
- 3D pore-graded PCD with MPL
- MEA based on commercially available reinforced membrane, coated with commercial Pt/C (cathode) and Ir/Aerogel catalyst

The initial results from the work packages (WP) on component manufacturing as well as compliance testing and protocols will provide the necessary feedback to optimize the requirements for production and performance.

## **4 Report on Manufacturing Process of PRETZEL stack components**

### **4.1 Planar bipolar plates (BPP) with corrosion resistant coating**

In order to separate the hydraulic medium from the inner cell components and to interconnect the individual cells of a high pressure PEMEL stack based on hydraulic compression, an optimized pole plate needed to be developed. In order to meet the requirements, the focus in pole plate development was on operability and long-term stability.

The stack design based on hydraulic single cell compression works with planar monopolar plates. Most important requirements are a high electric and a corresponding high thermal conductivity as well as a certain mechanical flexibility. A high thermal conductivity is required, since the hydraulic fluid is also used for temperature management and,

therefore, a good transition of the thermal energy from the process site to the hydraulic medium needs to be guaranteed. The electrical current is guided from one cell to another via several contact points. Although the in-plane distance between these points is being kept low, a homogeneous current distribution throughout the active area is favoured by a pole plate material with high in-plane conductivity. On the other hand, the material's thickness must not be too high, as the cell compression is provided by a hydraulic liquid surrounding the single cells and pole plates. Here, a flexible material distributes the hydraulic pressure equally to the active cell components, hence, guaranteeing for a homogeneous compression and with that homogeneous operation conditions.

To achieve high thermal and electrical conductivity, copper was chosen to be the base material. Different approaches for corrosion protection at the oxidizing environment present at the anode side of PEMEL were discussed in the consortium. An already tested simple solution with a thin Titanium foil was set to be the backup solution. The problem with this setup is the additional contact resistance between the foil and the copper pole plate, especially with oxide layers being present at the respective surfaces.

Therefore a corrosion protective layer needed to be developed for the copper pole plates, which keeps the electrical conductivity at a high level. This target could, for example, be reached by using a bilayer structure with Titanium for corrosion protection and e.g. Platinum for a low contact resistance. But since this structure is cost intensive, an approach without the need for precious metals was developed with Niobium as protective layer combining both desired properties for stability and conductivity. However, especially the possibility to withstand the harsh conditions at the anode side of PEM electrolyzers needed to be investigated. Nb-multilayer was applied by vacuum plasma spraying (VPS), and the coating showed significant stability. This has been validated by several corrosion measurements, which are described in detail in deliverable report D4.1 "Report of electrochemical evaluation of coated components" of the PRETZEL project.

VPS is a thermal spraying process, which enables the production of thick layers of several tens of microns with controlled porosity at scalable production rates. As spraying material various metallic and ceramic powders in the range of nanometres and hundreds of micrometres can be used. Furthermore, the plasma spraying technology is very suitable to produce dense coatings on low cost BPPs based for example on stainless steel substrate but also porous layers due to the mechanical stability and relevant thickness of the produced layers<sup>8</sup>. The main part of this technology is the torch, where the gas flows through the annular gap of a finger like cathode and a concentrically surrounding anode. The gas consists of a mixture of Ar, N<sub>2</sub>, H<sub>2</sub> and He and it becomes ionized by electric arcs between the electrodes of several hundred amperes and is heated up to at least 10,000 °C. The heat enables the complete or semi melting of the powder and accelerates it into the direction of the substrate. The plasma enthalpy, the powder injection technology and particle size of the material affect this process. A completely melted particle is ideal for producing dense and protective layers. Conversely, partly melted particles can be used for manufacturing porous coatings for multiple purposes. Therefore, 0.5 mm thick Cu plates were coated with 8 layers of Nb particles for the anode side pole plates. The used metal powder had particle size of 45 µm.

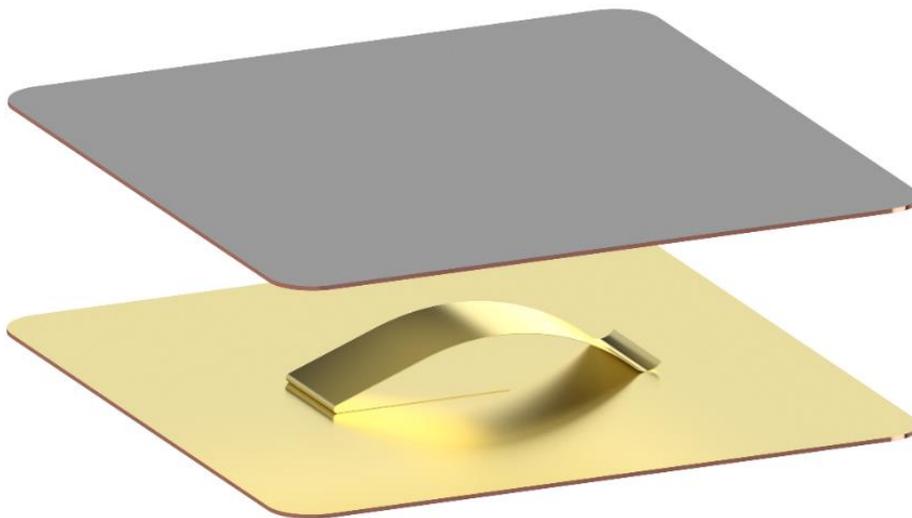
At the cathode side, gold coated copper plates are used. Gold coating is commonly used in the field of electrical connectors to lower the contact resistance. It is also used with hydrogen applications for a reduced hydrogen diffusion, e.g. as membrane coating for pressure sensors. It has, therefore, proven its applicability as coating for the cathode side pole plates. In contrast to electrical devices, where gold coatings are applied only to realize a low contact resistance, for the usage in PEMEL, it must also be assured that no material is present at the surface, which could negatively affect the process or the

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<sup>8</sup> Fauchais P. Understanding plasma spraying. J Phys D Appl Phys 2004;37:R86–108.

long-term stability. Therefore, a special three-layer coating is used in order to guarantee for the necessary stability and purity at the surface. After cleaning the copper plate, especially to remove the oxide layer, a nickel coating is applied. On top of that, a first gold layer is applied. The coating is done via galvanic methods. The electrolytes for these first two layers are designed to create a strong bonding between the different materials at the respective interfaces. At last, the so prepared pieces are plated with the final pure gold material. To ensure a long-term stable high-quality operation, gold coating is also applied on the outer side of each pole plate, both anode as well as cathode side.

Electrical connection between neighbouring cells is provided by flexible springs as shown in Fig 1. The springs need to meet the necessary specifications concerning the distance between the pole plates and the current load capacity. In order to keep the current density as homogeneously distributed throughout the active cell area as possible, current conduction from one monopolar plate to the next one needs to be established via as many connections as possible. Since a mechanically fixed connection of the pole plates would work against the principles of the hydraulic concept, a flexible solution was developed using springs. These springs are mounted to one side of each cell, contacting the backside of the neighbouring cell when stacked together. For long-term stability of the electrical contact, the springs are gold-plated similar to the pole plates.



**Fig 1.** Electrical connection between neighboring cells

The given dimensions of the pole plates and the springs allow for a maximum number of 85 springs being placed at the backside of the cathode pole plate. A small piece of glue, which is situated at the flat area on the underside of each spring, is used to attach the springs to the pole plate. When the cells are stacked together, the upper round shaped side is pressed against the next pole plate while the free end of the spring is in contact with the surface it's attached to. Electrical current is guided between neighbouring cells via these two contact points. Current conduction was investigated using the selected product in advance of the pole plate manufacturing process. The experimental study revealed that the Ohmic resistance of this kind of electrical connection amounts to about  $10 \mu\Omega$  per cell. This value corresponds to the usage of 85 springs per cell. Just like the pole plates themselves, the springs are gold coated for minimizing the contact resistance.

The final developed pole plate, hence, comes with the anti-corrosion protective layer faced to inner side of the cell and a flexible conductor on the outside, which is in contact to the hydraulic medium.

## 4.23D pore-graded porous current distributor (PCD) with macro porous layer (MPL)

The PCDs are the connecting part between BPP and the electrode of the MEA. Its importance is based on the good supply of the cathode electrode with electrons collected from the anode side electrode where water is split to oxygen and hydrogen and electrons. Secondly the PCD overtake the function of efficient water distribution and product gas removal to and from the electrode, respectively. Therefore PCDs need to fulfill the requirements of good electrical conductivity and low contact resistance which increase by lowering porosity. In contrary high porosities reduce limitations by the mass transport of water and gas through the structure. Regarding the material used for PCDs it needs to be highly corrosion resistant at potentials higher than 2 V in oxygen atmosphere.

Initially the production of the current collectors were planned to be done via two different techniques. On the one hand PCDs should be manufactured by selective laser melting (SLM) with gradient porosity. On the other hand the porous graded structures should be prepared by classical powder metallurgical (CPM) techniques. For both manufacturing routes samples were produced.

The PCDs were optimized according to the specifications defined in WP 2 "Definition of specifications and requirements" and manufactured in small scale as well as for the final stack design. The manufacturing process of the samples to be used in the PRETZEL stack is based upon the CPM technique. By improving this technique, a more homogeneous coating on the structural supporting material was generated.

Finally the PCDs in stack dimensions (500cm<sup>2</sup>) were manufactured via CPM technique: By deposition of a fine Ti powder layer on the coarse support structure followed by a heat treatment (sintering), an asymmetric porous structure is generated.

Sintering is needed for diffusion bonding of the powder particles and joining the MPL to the support structure. Typical sinter temperature is 70% of the absolute melting temperature. The optimal sinter temperature varies also within the particle size distribution of the metal powder: the finer the powder the higher the diffusion activity. The batch furnace used for heat treatment is build up with metal heating elements and metal shielding without any graphite elements to prevent carbide formation. The heat treatment is performed under a vacuum of  $2 \cdot 10^{-5}$  mbar.

The finally used substrate for the PCDs is an expanded metal with a density of about 2 g cm<sup>-3</sup> that means consequentially a porosity of about 55 %. These metal sheets were made by cutting slits into a plate and stretching this plate afterwards. This process creates a specific pattern with open gaps. To produce the desired pore graded structure the fine powder was deposited on the aforementioned substrate to generate the macro porous layer (MPL) by sintering in high vacuum to prevent oxide formation during heat treatment. This specific layer enables the reduction of contact surface resistance as well as the mass transport limitation by optimizing the pore sizes in contact to the electrode. MPL requirements such as thickness, porosity, particle size, pressure drop, electrical conductivity, tolerances, etc. were also defined in WP 2. For all MPL the same fine powder fraction was used for the standard powder metallurgical process. An image of a supplied expanded metal sheet compound is exemplary shown below in Fig. 2.



**Fig. 2.** Delivered Ti PCD made by CPM for partners

Several rounds of evaluation with work packages regarding definition of specifications and requirements as well as compliance testing and protocols led to an optimized coated PCD in terms of efficiency, costs and performance in particular at the challenging operation conditions of PRETZEL project.

### **4.3 Membrane electrode assembly (MEA) based on catalyst coated membrane (CCM) manufacture**

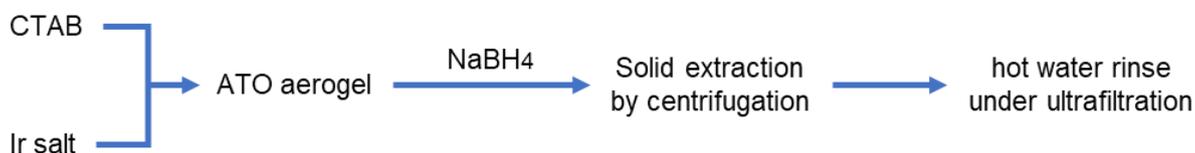
One of the core components of each electrolyzer cell is the MEA which is in contact with PCDs on anode as well as on cathode side so that current can flow to and from the electrodes. Good membranes are distinguished by high proton conductivity, mechanical stability and low gas permeability. Additionally, the acidity of the membrane together with the oxidative environment forces the use of precious metal electrodes. Regarding cost, lifetime and efficiency of PEMWE especially the choice of anode catalyst has a significant impact. Commonly Pt is used on the cathode for the hydrogen evolution reaction (HER) while Ir is the state-of-the-art anode electrocatalyst for the oxygen evolution reaction (OER). Due to the high price, low annual production value and the fact that Ir is one of the rarest element in the earth crust large-scale introduction of PEMWE technology will be limited. Therefore, a reduction of catalyst loading is urgently needed. Either highly active Ir-based catalysts need to be developed or electro-conductive ceramics as substrate materials could be used. Regarding the latter approach nano-sized iridium supported on SnO<sub>2</sub>:Sb aerogel catalysts were developed in advance to the project and showed high activity concurrent with excellent stability. Due to a catalyst reduction of 70% while keeping same OER activity this strategy was pursued for PRETZEL stack application.

Antimony doped tin oxide, SnO<sub>2</sub>:Sb (ATO) aerogel was synthesized through sol gel process starting from alkoxide precursors. First of all two solutions were prepared: Solution A was prepared diluting 1.6 mmol of Sn(OiPr)<sub>4</sub> in isopropanol and the required amount of the dopant agent. Solution B was prepared by diluting 0.058 ml of HNO<sub>3</sub> and 0.038 ml of water in isopropanol. Then solution B was added dropwise to solution A, and after the gelation the gel was covered with isopropanol. After 3 cycles of washing the gels, these were dried with supercritical CO<sub>2</sub>. Once the gels were dried, they were calcined at 600 °C for 5 h.

Modified synthesis following Lettenmeier et al. (see Fig. 3) was selected for the preparation of the anode electrocatalyst. The procedure consists in an impregnation on ATO support suspension of a mixed solution of the iridium salt precursor (IrCl<sub>3</sub>) and capping agent (CTAB) and further reduction with NaBH<sub>4</sub> under argon atmosphere and stirring.

The solvent used in all cases was anhydrous ethanol and the solid was extracted by centrifugation in conditions of 7400 rpm for 4 minutes. Further the solid was submitted to ultrafiltration and rinse repeatedly with hot water. Then the catalyst was achieved by drying under argon atmosphere at room temperature.

**Lettenmeier et al** preparation (argon, anhydrous ethanol):



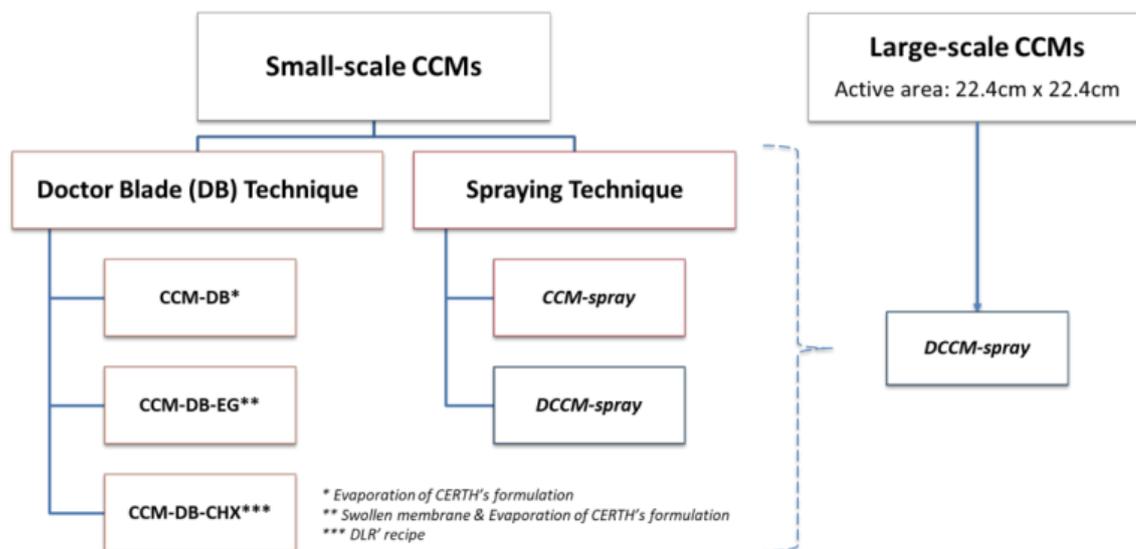
**Fig. 3.** Procedure of catalyst synthesis reported by Lettenmeier et al.

After synthesis of sufficient amount of ceramic aerogels to be used as catalyst substrates (characterization performed in WP 4 “Compliance testing and characterization”), IrO<sub>x</sub>-Ir catalyst supported on the developed substrates was prepared. Two (2) formulations for the supported catalysts were targeted: 30% Ir on ATO and 70% Ir on ATO. Moreover, commercial ATO purchased from Sigma Aldrich was also used as support material for comparison reasons.

To prepare MEAs targeting to current density of 4 A cm<sup>-2</sup> (6 A cm<sup>-2</sup> peak current density) at potentials below 2 V and under pressurized PEM electrolysis conditions different approaches were considered. The MEA manufacturing needs a strong optimization of the manufacturing process by tuning the conditions (pressure, temperature, pressure time) in order to match the properties of all the components. To optimize the manufacturing process, a development plan was created and two CCM techniques were applied to manufacture MEAs:

- a) through spray coating
- b) using a film applicator system (doctor blade).

Commercially available single-sided ELAT electrodes were employed as cathode materials and hot pressing of all individual components integrated the MEAs manufacturing procedure for “single” CCMs, meaning only the anode side is a catalyst-coated-membrane. As a following development step, attempts were focused on the CCM technique for the application of the Pt/C electrocatalyst at the cathode side, through spray coating directly on the membrane (double-CCMs – DCCMs). Fig. 4 illustrates the routes which were followed for optimization of CCMs manufacturing and the selection of the technique for large-scale CCMs manufacturing.



**Fig. 4.** Techniques of CCM manufacturing

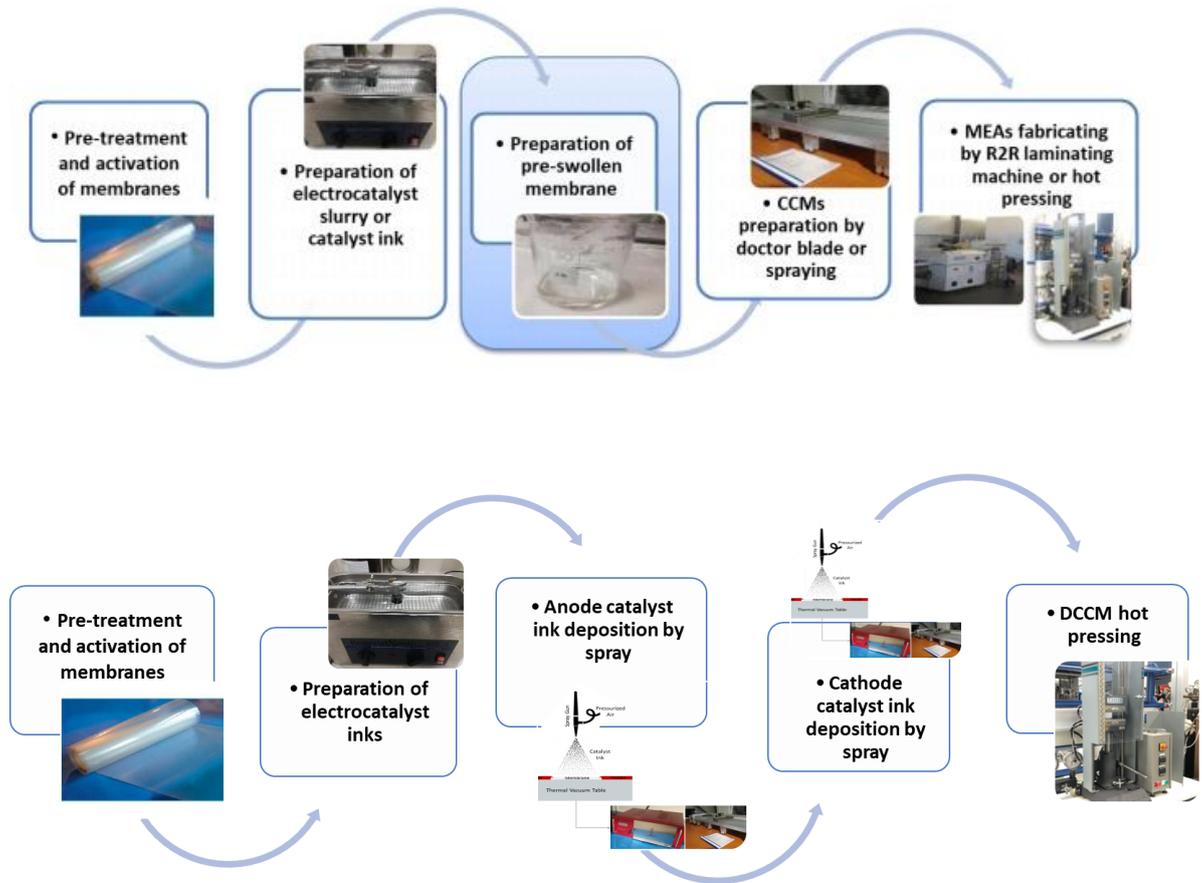
During the development of the manufacturing route, 27 batches of MEAs were prepared in total leading to the conclusion that sprayed DCCMs should be used for implementation to the PRETZEL stack.

Since spraying technique is currently the standard procedure that can be industrialized via mainly robotization, a great deal of effort has been put into mastering the spraying techniques of the CCM on a small-scale.

The development was focused on MEAs of anode and cathode side prepared both with the CCM technique. This development is expected to deliver optimum catalyst adhesion on the membrane, thus eliminating undesired interfaces and consequent undesired resistances, while maximum catalyst utilization (low loading-high accessibility) is also achieved. This approach aimed to be optimized in the small-scale MEAs and afterwards to be applied in the large scale MEAs for the stack. Various batches of double-CCMs with reference material, IrO<sub>2</sub> and electrocatalysts from PRETZEL project (30% Ir/ATO(SA), 30% Ir/ATO(ARM) & 70% Ir/ATO(ARM)) were manufactured. The electrocatalysts containing 30% and 70% Ir supported on ATO, were produced by the synthesis method reported by Lettenmeier et al.<sup>9</sup>, as it was proven to be the most promising among other synthesis routes evaluated. The catalysts employed for the MEAs preparation at loadings of 1 mg/cm<sup>2</sup> and 0.3 mg/cm<sup>2</sup> (Ir-based) and 0.2-0.4 mg/cm<sup>2</sup> (Pt-based).

The selected manufacturing route followed the steps displayed in Fig. 5.

<sup>9</sup> P. Lettenmeier, S. Kolb, F. Burggraf, A.S. Gago and K. A. Friedrich, Towards developing a backing layer for proton exchange membrane electrolyzers, J Power Sources, 2016, 311, 153-158, DOI:0.1016/j.jpowsour.2016.01.100



**Fig. 5.** Procedure and method for DCCM manufacturing

As a first step, the pre-treatment and activation of the membranes before use carried out.

The second step of the MEA manufacturing process described in Fig. 5 is the preparation of the electrocatalyst ink. For the DCCMs the anode catalyst ink was prepared by mixing the proper amounts of the anode catalyst powder ( $\text{IrO}_2$  commercial (Alfa Aesar) or 30% Ir/ATO (Sigma Aldrich) or 30% Ir/ATO (ARMINES) or 70% Ir/ATO (ARMINES)), Nafion solution 5% w/w in water and alcohols, isopropanol/double distilled water. The ink is sonicated in an ultrasonic bath, until it becomes homogeneous. Cathode catalyst ink was prepared by mixing the proper amounts of the catalyst powder (Platinum, nominally 40% on carbon black, HiSPEC® 4000 (Alfa Aesar)), isopropanol, double distilled water, Nafion solution 5%) and is sonicated in an ultrasonic bath, until it becomes homogeneous.

As third and fourth step the spraying of anode and cathode electrocatalyst inks on the membrane produces a single and double CCM, respectively. Coating of the anode electrocatalyst is achieved through spraying of the catalyst ink on the membrane, which is deposited on a thermal vacuum table at a constant temperature. Vacuum is needed in order to prevent swelling of the membrane during the spraying procedure and have a homogeneous catalyst layer. Now a single-CCM is produced. By coating of the cathode electrocatalyst which is achieved through spraying of the catalyst ink on the back side of single anode-CCM, a double-CCM is produced. The single anode CCM is deposited on a heated vacuum table at a constant temperature.

Finally, the DCCMs are hot pressed in a hydraulic press (Carver 3851) at  $130^\circ\text{C}$ .

## 5 Conclusions

Within the game changer project PRETZEL, major step improvements in terms of cost reduction, performance and efficiency are delivered. It is actually expected to surpass perspective the FCH JU's KPI targets in terms of cost, efficiency, lifetime and operability with further development based on this project by the year 2023. PRETZEL addresses all suggested points of improvement and aims to demonstrate the improvements of the single components as well as the interaction of the innovations in a 25 kW scale high pressure system.

In this report, details of the BPP, 3D-pore graded PCD and MEA manufacturing routes are presented. The developed component designs guarantee for a long-term operation at the challenging "game changer" operation conditions of 90°C, peak current density of 6 Acm<sup>-2</sup> and 100 bar. Each component ensures high efficiency while cost is reduced.

For the BPP this is realized by minimized contact resistances at the interfaces to the PCDs as well as between neighbouring cells while assuring high durability. The planar structure of the pole plates allows for an easy handling regarding the corrosion protective coatings, which are applied by different techniques. The corrosion resistance of the Nb-coating was electrochemically evaluated and demonstrate absolute resistivity against the aggressive surrounding.

The pore-graded PCDs manufactured via classical powder metallurgical techniques on titanium mesh-type substrate outperform all comparative electrochemically measured PCDs regarding performance and durability. Therefore, after the intensive physical characterization as well as electrochemical testing the aforementioned samples were chosen for final PRETZEL stack application.

The CCM development was optimized to select the appropriate technique for large-scale MEA manufacturing. PEMWE measurement (single cell performance evaluation under water electrolysis) as well as based on processes and results (quality and reproducibility in preparation of electrodes and MEAs and their performance), the sprayed double-CCM approach was selected for application to the PRETZEL stack in the project.

All samples were tested according to our compliance testing protocols and analytics described in the public deliverable D2.2 "Compliance test protocols and analytics". Within several iteration processes the described components were optimized to reach the proposed overall requirements needed to fulfil the project targets. With these advanced components deeply evaluated the final stack operation in the large high-pressure stack is highly promising in reaching the project goal of a long-term test.

## 6 Appendix