



**CRITICAL RAW MATERIAL ELECTROCATALYSTS REPLACEMENT
ENABLING DESIGNED POST-2020 PEMFC**

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DELIVERABLE REPORT

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SUMMARY**Keywords**

Components, MEA and performance requirements

Abstract

In this deliverable report the technical specifications for the CRESCENDO single cell components are presented. The specifications are based on the automotive requirements and operating conditions. In addition standard testing protocols and sensitivity tests are defined for the purpose of fair comparison between partners, while integrating European harmonised protocols where relevant.

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1. INTRODUCTION

This deliverable describes cell requirements as well as testing protocols. The performance test will be carried out according to the test protocols defined in WP2 on $5\text{cm}^2_{\text{geo}}$ up to automotive size MEA. The tests will include parameter variation evaluating the sensitivity to operating temperature, pressure, humidification, and stoichiometry.

2. SCOPE

The aim of this report is to compile the technical specifications for the CRESCENDO single cell and potential short stack. Initial estimation of a full scale stack requirements will be conducted based on estimation of maximum capacity of non-PGM based electrodes. The specifications are based on the automotive requirements and operating conditions also provided by BMW in this deliverable D2.1. This deliverable report identifies the operation, performance and material requirements (MEA, GDL, PEM and bipolar plate) as well as the final targets for the project. In addition, this documents specified the standard testing protocols, integrating European harmonised protocols where relevant, for a fair comparison of catalysts and CCMs between project partners and between FCH projects. It has to be noted that at this stage the D2.1 document serves as a starting point and it is expected that it will be revised along the project based on the project's results.

3. DISCUSSION

3.1 COMPONENTS SPECIFICATION

a. Metallic Bipolar Plate Specification

A bipolar plate (BPP) is a major stack component, which purpose is to deliver both reactant gases and the coolant into specified media paths. An anode half-plate and a cathode half-plate together define the 3 media flow pathways (oxidant, fuel and coolant). In addition, the bipolar plate provides the cell voltage measurement points, as well as positioning features, stack assembly features, half circular identification marks and part markings, which facilitate stack assembly.

A bipolar plate consists of a metal stamped anode and cathode plate joined together by laser welding to form a tight assembly. A conductive coating and an integrated bead seal with a thin layer of sealant material on top are integrated into the BPP module. The base material of the anode and cathode plate is ANSI 316L stainless steel. The bipolar plate that is going to be used within this project is developed within FCH 2 JU INSPIRE. During the course of INSPIRE, several upgrades will be made to the bipolar plate design (sealing, flow field geometry etc.). The structure of the plates, the flow field, ports, distribution zone and all geometric features are designed according to the requirements to fulfil the stack operating conditions specified within INSPIRE project.

At the moment the INSPIRE bipolar plate design has not reach the necessary maturity, and during the course of the CRESCENDO project, a new generation of flow field design will be presented and used. Therefore, defining the MEA and bipolar plate mechanical interface can only be done for the current

design of bipolar plate-table 1. This table will be updated and adapted based on the progress within INSPIRE, during the course of the CRESCENDO Project. Table 1 is therefore to be used only as a starting point for discussion.

Table 1: Mechanical interface tolerance for MEA and Inspire bipolar plate

Specifications	Values	Unit
Active area of MEA	287,5	cm ²
x-value active area of bipolar plate	250	mm
z-value active area of bipolar plate	155	mm
Dimension in y-direction at 2 MPa	0,350	mm
Overlap of CCM with GDL	3	mm
Deviation of clamping force at active area	0,05	MPa
Min clamping force	1	MPa
Max. clamping force	2,5	MPa
Max. continuous pressure difference	1	Bar

b. Gas Diffusion Layer Specification

Gas diffusion layers (GDLs) consist of a carbon fibre-based supporting substrate (GDL backing) coated with a micro-porous layer (MPL). In the cell assembly, the GDL substrate is facing the bipolar plate and the MPL is in contact to the catalyst layer. With respect to the integration into the MEA and the cell, the most important parameters of the GDL are the electrical and mechanical properties as well as the pore characteristics of the GDL. Narrow tolerances in thickness and compressibility are required in order to reduce channel intrusion, electric contact resistance, as well as to ensure compatibility with the sealing type and its thickness. Depending on the targeted operating conditions within CRESCENDO, the GDL design parameters are going to be selected. The suggested range is described in Table 2. At this stage most parameters are unknown as no data are available yet regarding the performance and water management requirements of non-PGM catalyst layers.

Also, the GDL thickness cannot be defined at this stage of the project, as it will depend on the catalyst layer thickness. This is due to the restriction of the total MEA thickness (<370µm) originating from the height and the compressibility of the bead seal of the bipolar plate. Based on the typical clamping force applied on the cells, the GDL is compressed between 20-27%. Once the catalyst layer has been optimised and the thickness of the electrodes is defined, the GDL thickness will be calculated by subtracting the incompressible thickness of the catalyst layer and the PEM by taking also into consideration the height of the sealing. Table 2 presents a suggestion of GDL properties to be used as a starting point. During the course of CRESCENDO, table 2 will be revised and adapted to the GDL which will fulfill the performance and geometric requirements of this project.

Table 2: Gas diffusion layer specifications - range of acceptance

Specifications	Values	Tolerances	Unit
Thickness of GDL(uncompressed)	tbd	± 15	µm
Thickness of GDL (compressed @ 1MPa)	tbd	± 15	µm
GDL area weight	tbd	± 5	g/m ²
MPL loading	tbd	± 5	g/m ²
Backing porosity in %	>85	-	%
PTFE content of MPL	tbd	± 5	wt %
Bending stiffness X/Y	>1,5/ >1,0	-	Nmm
TP gas permeability (uncompressed)	2,5*	± 0,5	10 ⁻¹² m ²
IP gas permeability (uncompressed)	4,7	± 0,5	10 ⁻¹² m ²
IP gas permeability (compressed @ 1 MPa)	1,8	± 0,5	10 ⁻¹² m ²
TP electronic resistance (@ 1 MPa)	<10	-	mΩ cm ²

*Calculated from Gurley measurement

c. Polymer Electrolyte Membrane (PEM)

The heart of a PEM fuel cell is a polymer membrane that presents specific capabilities. A suitable polymer electrolyte membrane needs to fulfill the following requirements: high proton conductivity, very low gas permeability of fuel and reactant gases and high chemical and mechanical stability in the demanding fuel cell environment. The state of the art membranes consists of perfluorocarbon sulfonic acid ionomers (PFSA). Typically for automotive application the membrane thickness is below 20 µm, with a reinforcement to ensure high mechanical stability. Table 3 represents the desired properties of the membrane for automotive applications.

For the initial stages of the CRESCENDO project JMFC will provide a 15 µm thick PFSA membrane. Based on the progress and performance of the MEAs, it might be needed to re-evaluate the type of PEM used in order to achieve the performance and durability targets.

Table 3: Polymer Electrolyte Membrane Specification

Specifications	Values	Unit
H₂ crossover (30-100% RH and 50-90°C)	< 2	mA/cm ²
Conductivity	> 0.1	S/cm
Swelling at 100% RH	< 30	%
Tensile strength at ambient conditions	>40	MPa
Elongation at break at ambient conditions	>150	%
Minimum Temperature stable	100	°C

d. Membrane Electrode Assembly Specification

A membrane electrode assembly comprises: the polymer electrolyte membrane, anode and cathode catalyst layers; a polymeric sub-gasket frame material as well as anode and cathode gas diffusion media. Typically, the catalyst layers and membrane are laminated by a hot pressing procedure, under specific

humidity and temperature conditions based on the type of components, to form a catalyst coated membrane (CCM). Thereafter the catalyst coated membrane is sealed between a polymeric sub-gasket frame via a hot bonding step. Finally the GDL is adhesively bonded to the assembly to form the membrane electrode assembly. This process is slightly altered when gas diffusion electrodes (GDE) are used.

The overall MEA thickness compressed under 1 MPa needs to fulfil specific requirements and is restricted by the height of the sealing material on the bipolar plate. As specified in previous sections, the sealing height will change over the course of the Inspire project. Testing MEAs in automotive size cells is scheduled to start in M24 within CRESCENDO. Thus by M18, the latest and upgraded bipolar plate design will be available. Once the bipolar plate specifications are known table 4 can be updated, and hence the total thickness of the MEA can be determined. The maximum possible MEA thickness cannot exceed 370 μm .

Table 4: Thickness tolerance for MEA @1 MPa

Specifications	Values	Tolerances	Unit
MEA thickness (compressed @ 1MPa)	<370	± 20	μm
Cathode electrode thickness	tbd	-	μm
Anode Pt electrode thickness	5	± 1	μm
Anode non-PGM electrode thickness	tbd	-	μm
Seal subframe of MEA	90	± 10	μm

The electrochemical performance specification is shown in Table 5 at the important operating points for the single cell. Table 5 contains an initial target for operating conditions. During the course of CRESCENDO, the newly developed electrodes will be subjected to temperature, RH and pressure sensitivity tests in order to determine the optimum operating conditions for these newly developed catalyst and catalyst layers. Table 5 serves as a starting point for this investigation and might need to be altered during the following months, while Table 6 shows the MEA loading specification and target power density.

Table 5: Target points for electrochemical performance of the BMW MEA single cell

Specifications	Unit	Mode 1	Mode 2	Mode 3	Mode 4
Current density	A/cm ²	0,1	1,4	0,6	0,6
Required cell voltage*	V	0,75	0,3	0,71	0,7
Cell temperature in	°C	65	90	90	80
Anode					
Pressure anode inlet	Bara	2,3	2,3	2,3	2,5
H2 concentration anode (dry)	mol%	100	100	100	100
N2 concentration anode (dry)	mol%	0	0	0	0
Dew point anode inlet	°C	45	65	65	63,8
RH anode inlet	%	38,5	35,7	35,7	50
Stoichiometry H2		1,4	1,4	1,4	1,3
Cathode					
Pressure cathode inlet	Bara	2,3	2,3	2,3	2,3
O2 concentration cathode (dry)	mol%	21	21	21	21
N2 concentration cathode (dry)	mol%	79	79	79	79
Dew point cathode inlet	°C	45	65	65	53
RH cathode inlet	%	38,5	35,7	35,7	30
Stoichiometry Air		1,8	1,8	1,8	1,5

Table 6. MEA loading specification and target power density and durability for the end of the project

Specifications	Range Values	Unit
Anode Pt loading	0,025- 0,1	mg Pt/cm ²
Anode non-PGM	tbd	mg/cm ²
Cathode catalyst loading	1,0 - 4,0	mg/cm ²
Target power density (@ 0,7 V)	0,42	W/cm ²
Target power density (@ 0,6 V)	0,50	W/cm ²
Performance loss H2/Air after 1000 h @ 1,5 A/cm²	< 30	%

Table 7 contains the stack requirements based on the performance targets set by this project. Determining the stack requirements containing PGM-free based MEAs will help in the cost analysis to establish the feasibility in automotive application (D.2.4).

Table 7. Stack requirements based on performance targets of Table 5

ID#	Stack requirements: Specifications	Value	Unit
Electrical Requirements			
1_1	Stack power @0,6 V	56,7	kW
1_3	Stack maximum voltage	<800	V
1_4	Maximum operation cell voltage	0,85	V
1_5	Cell to cell voltage variation at any operating condition	+ - 10	mV
1_6	Maximum current at peak power	600	A
1_7	Maximum current at normal power	<600	A
1_8	Maximum dynamics of current	1000	A/s
1_9	Maximum Cell Count	394	#
Cooling Requirements			
1_10	Stack maximum outlet temperature	95	°C
1_11	Maximum pressure drop cooling loop	575	mbar
1_12	Cooling liquid	50/50	%/ % Glycol/DI- Wasser
1_13	Coolant conductivity	<20	µS/cm
Cathode Requirements			
1_14	Stack cathode maximum outlet pressure	2	bara
1_15	Maximum cathode pressure drop	525	mbar
1_16	Cathode stoichiometry min(lambda)	1,3	-
1_19	Cathode stoichiometry max(lambda)	1,8	-
Anode Requirements			
1_20	Maximum anode pressure drop	200	mbar
1_21	Anode stoichiometry min(lambda)	1,25	-
1_22	Anode stoichiometry max(lambda)	1,5	-
1_23	H2 quality	SAE2719	-
1_24	Minimum H2-concentration inlet; dry	83,7	mol% H2

3.2 SUMMARY OF TESTING SPECIFICATION

Our testing protocols for laboratory and automotive size cells will be based on EU harmonised conditions for PEMFC testing [1]. Details of the testing protocols are described in detail in Appendix 1. The sensitivity tests described in section 3.25.-3.2.8 will be conducted only on the most promising samples.

3.2.1 Control parameters

Fuel cell test benches gives us the option to control various parameters using inlet or outlet value from the system. If possible, it should be agreed between all partners from this consortium the way to control these parameters. The controlled parameters are:

- Inlet or outlet gas pressure (anode and cathode compartment)
- Inlet or outlet cell temperature (anode and cathode)

It has to be noted, that not all cells use a liquid cooling/heating medium to control temperature of the fuel cell. In the case that heating resistors are used, the thermocouple should be placed in the middle of the cell monopolar plate in both anode and cathode to monitor the temperature of the cell.

3.2.2. Cell hardware configuration

Different cell hardwares are available between partners of the consortium. The active area varies between $5 \text{ cm}^2_{\text{geo}}$ and $50 \text{ cm}^2_{\text{geo}}$ for the laboratory scale MEAs up to $\sim 300 \text{ cm}^2_{\text{geo}}$ for the automotive size MEAs. The coolant flow and reactant flow (fuel /oxidant) should be counter flow in all the above mentioned hardwares.

3.2.3. Leak test

The objective of the leakage test is to determine the leakage rate decrease of the anodic and cathodic compartment and assure the gas tightness of both the cell and test stations. The leakages is separated in internal and external leakages. The external leakage procedure is specified by the EU harmonized Protocol and should be followed by all partners of the consortium before each test.

As PGM-free cathode are going to be used in this project, determination of the H_2 crossover via the conventional way of measuring electrochemical H_2 oxidation rate (set by diffusion across the membrane) at cathode is not possible due to inactivity of non-PGM for H_2 oxidation, an alternative solution is to determine the internal leakage of the PEM. The following procedure is proposed for the internal leakage test:

- Purge with dry N_2 both sides of cell at minimum flow rate
- Pressurize the anode compartment by 2.15 bara, while keeping the cathodic compartment pressurised at 1.15 bara.
- Reduce the N_2 flow in both compartment to zero
- Keep cell at this state for 10 min
- Calculate the internal leakage by the increase of the pressure in the cathodic compartment with time

Acceptance criteria for external leakage lower than 7 mbar/min, while internal leakage lower than 1 mbar/min. Although contributions from hydrogen crossover are not expected to be significant with the use of non PGM cathodes, state of the art protocols (references listed in the document) will be used to verify that the performance obtained with the non PGM cathode is not influenced by the crossover of the reference Pt/C anode catalyst or hydrogen, in particular before and after durability testing. Nevertheless different H₂-crossover protocol will be investigated to see which is more suitable [2,3].

3.2.4. Polarisation curve

Polarisation curve is the main tool to characterize the performance of the MEA. The objective is to be able to determine the voltage and power density at a specific current density under controlled operating conditions. In order to be able to compare data between the different partners the EU harmonised Polarisation protocol should be used [1]. Appendix 1, Table 1 shows the EU harmonized polarisation curve protocol. For comparison reasons between different samples the polarization curve will be recorded under the conditions specified in Mode 4 in Table 5. This particular Polarisation protocol has been optimized for a PGM based MEA, so it might be adjusted to the PGM-free based MEAs if required.

3.2.5. RH sensitivity

The effect of the relative humidity of the gas inlet for both anode and cathode on the cell performance have to be investigated. Since there are two sources of water during fuel cell operation, (1. Introduction of water vapour by the reacting gasses while passing through a humidification system and 2. The produced water due to the electrochemical reaction), a delicate balance must be achieved in order to obtain optimum performance.

For this reason, 4 polarisation curves will be obtained at a cell temperature of 80 °C, while changing the relative humidity of both anode and cathode from 30%, 50%, 70% and 100% RH. The gas stoichiometry and pressure should be defined by all partners.

3.2.6. Temperature sensitivity

The cell temperature influences both the performance and the long term durability of the single cell. The purpose is to evaluate the temperature sensitivity of the MEA when operating at low, medium and high temperatures. High operating temperatures have beneficial effects, such as higher kinetics and better water management. However under these conditions the MEAs might exhibit undesired effects such as, higher degradation rates and H₂ crossover. The following temperatures will be used for the temperature sensitivity test:

-low: 45 °C

-medium: 80 °C

-high: up to 95 °C (depending of the capability of the test benches of each partner)

The stoichiometry of reacting gasses and the inlet/outlet pressures will be defined by all partners.

3.2.7. Anode and cathode stoichiometry sensitivity

The cathodic electrode performance exhibits a dependence on the oxidant stoichiometry. Increase in the oxidant stoichiometry can change the water content inside the cell and thus affecting the proton conductivity of the membrane and in the catalyst layer. However low stoichiometries at high current

densities might lead to flooding of the electrode. For this reason it is useful especially for the development and optimisation of the cathode catalyst layer to identify the stoichiometry sensitivity.

Since in this project PGM-free anodes are also going to be developed, H₂ stoichiometry sensitivity should also be defined. The proposed stoichiometry range for the H₂ sensitivity tests 1,25-1,8 , while keeping the cathode stoichiometry at 2,0. While, the proposed stoichiometry range for the Air sensitivity tests 1,3-2,0 ,while keeping the anode stoichiometry at 1,8. The cell temperature and inlet gas pressures will be defined by the project partners.

3.2.8. Pressure sensitivity

The inlet gas pressure affects the fuel cell performance as it influences the OCV, the partial pressure of reacting gasses and water, H₂-crossover and mass transport resistance, to name a few. Generally increase in the reactant gas pressure increases the cell performance, however this effect is more enhance for the cathode due to sluggish ORR reaction. It is suggested that different polarization curves should be obtained at a specified cell temperature and fuel/oxidant stoichiometry in order to evaluate the effect of the gas pressure to the MEA performance. The pressure range for both anode and cathode would be 145 kPa_{a,inlet} up to 250 kPa_{a, inlet} .

3.2.9. Fuel and oxidant composition

In order to ensure accurate and reproducible polarization curves, the oxidant and fuel composition should assure low levels of contaminants that could reduce the MEA performance. As suggested by the EU Harmonized testing protocol, the hydrogen should be supplied in gas cylinders with quality grade of 5.0 in order not to influence the performance and the lifetime of the MEAs. Whereas for the oxidant, air should be oil free and filtered for dust particles according to ISO 85731: 2010. In addition for pure O₂ gas it should be supplied in gas cylinders with a grade of 5.0.

4. CONCLUSIONS AND FUTURE WORK

The initial components and MEA requirements are defined. The performance targets are set and the testing protocols are clearly defined. However the components specification will be reviewed during the course of this project in order to achieve the performance and durability targets.

5. REFERENCES

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6. APPENDIX – MEA TESTING PROTOCOLS FOR WP2

POLARISATION TESTING PROCEDURE: THE OPERATING CONDITIONS OF THIS POLARISATION CURVE ARE SPECIFIED IN TABLE 5.

Polarisation curve set-points

Set point no.	Current density A/cm ²	Recommended dwell time (s)	Recommended Data acquisition time (s)
1	0.00	≤60	≥30
2	0.02	≤60	≥30
3	0.04	≤60	≥30
4	0.06	≤60	≥30
5	0.08	≤60	≥30
6	0.10	≤60	≥30
7	0.20	≥120	≥30
8	0.30	≥120	≥30
9	0.40	≥120	≥30
10	0.60	≥120	≥30
11	0.80	≥120	≥30
12	1.00	≥120	≥30
13	1.20	≥120	≥30
14	1.40	≥120	≥30
15	1.60	≥120	≥30
16	1.80	≥120	≥30
17	2.00	≥120	≥30
18	1.80	≥120	≥30
19	1.60	≥120	≥30
20	1.40	≥120	≥30
21	1.20	≥120	≥30
22	1.00	≥120	≥30
23	0.80	≥120	≥30
24	0.60	≥120	≥30
25	0.40	≥120	≥30
26	0.30	≥120	≥30
27	0.20	≥120	≥30
28	0.10	≤60	≥30
29	0.08	≤60	≥30
30	0.06	≤60	≥30