

Disruptive pemfc stack with nOvel materiaLs, Processes,
arch**H**itecture and optimized **IN**terfaces

DOLPHIN Workshop, Ulm June 16th 2023

Technological highlights: components, production technology,
performance results

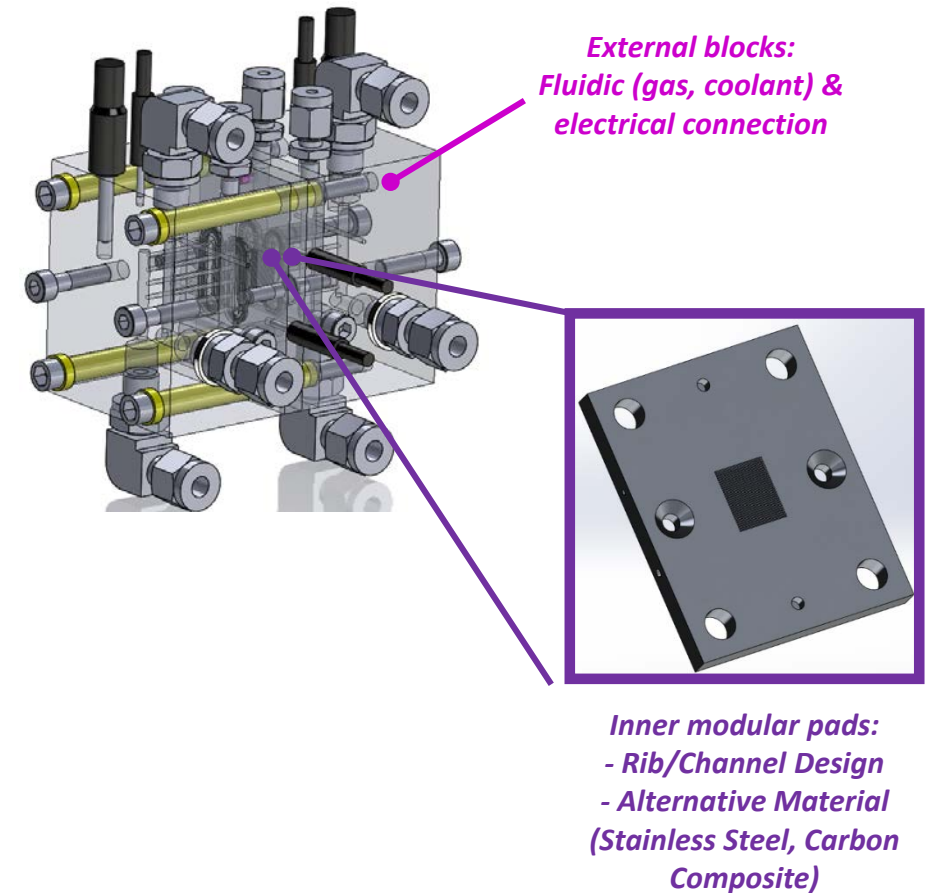
CEA Team : Fabrice MICOUD, Thomas CAVOUE, Clémence MARTY, Charlène SIGUENZA, Ludovic ROUILLON, Denis TREMBLAY,
Jean-François BLACHOT, Jean-Philippe POIROT-CROUVEZIER and Joël PAUCHET

ZSW Team : Benjamin WIEDEMANN, Michael SCHMID, Sepehr SAADAT, Florian WILHELM

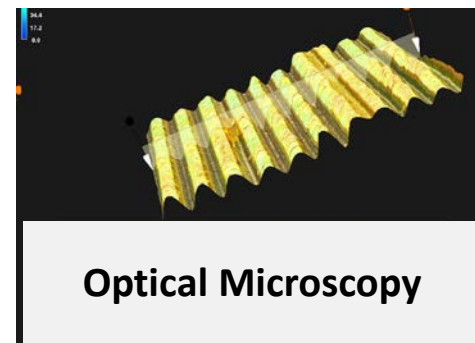
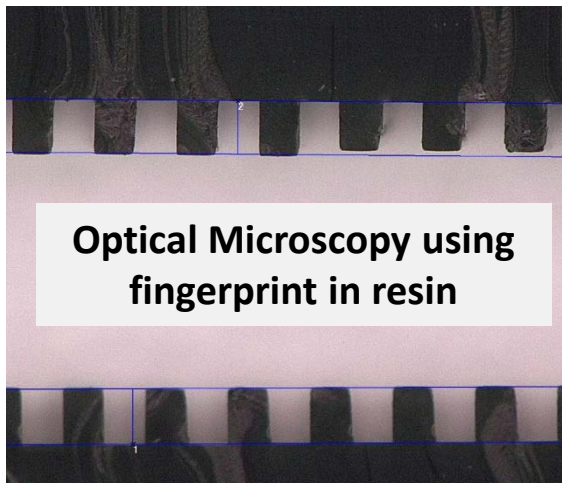


First evaluation of the different concepts EC and EFC at small scale

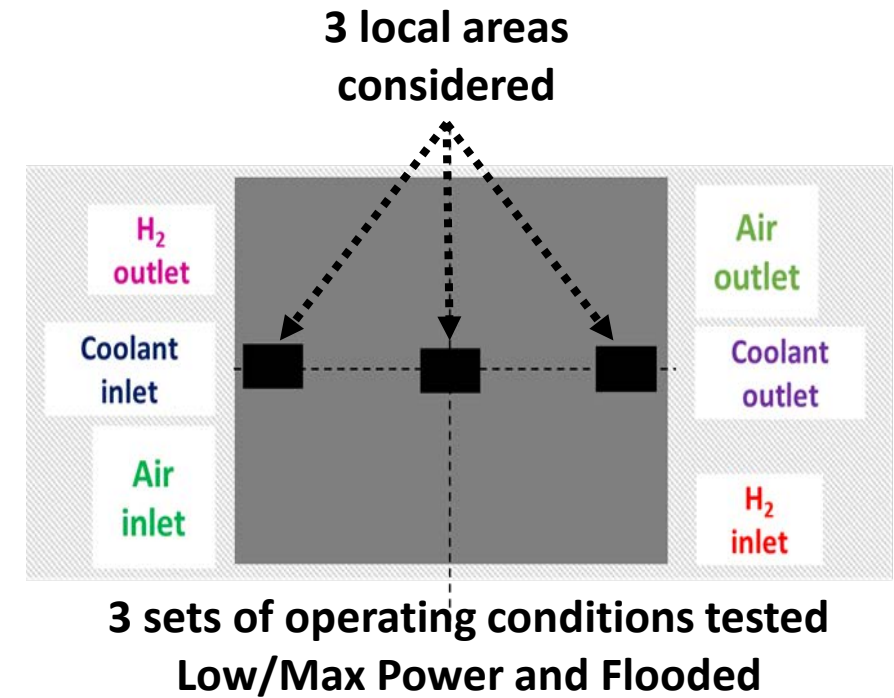
- **Use of differential cell « zero gradient »**
 - Homogeneous fluidic conditions
 - Homogeneous T°, RH and concentrations
 - Reproduction of local stack operation
- **Many designs and configurations to be evaluated**
 - Model Rib/channel designs (EFC)
 - ✓ 3D printing (CEA) and additive manufacturing (DMG Mori)
 - ✓ Channels formed into GDM
 - EC|EFC Interface
 - ✓ Optimisation of MPL layer (ZSW)
 - ✓ Removal of GDM/GDL
 - CCM material (membrane and electrode composition) (EC)
 - Impact of Single Layer Graphene onto the PFSA membrane (EC)
 - ✓ Presentation of SLG by University of Manchester



- Approach based on system and stack specifications
- Further definition of local operating conditions based on modeling at cell level
- Reference and commercial materials used as EC and EFC
 - Commercial Gore CCM (0.1 / 0.4 mg_{pt}/cm²) & Commercial GDL materials
 - Model flowfield designs by machining

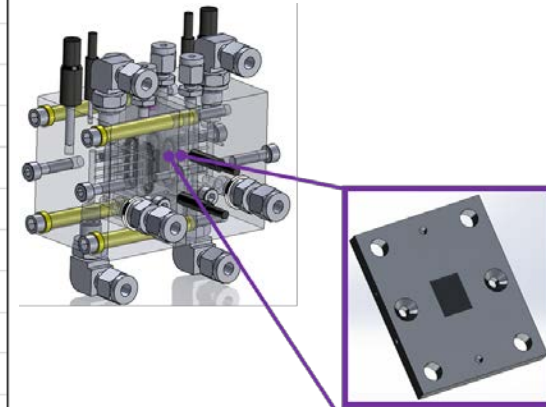
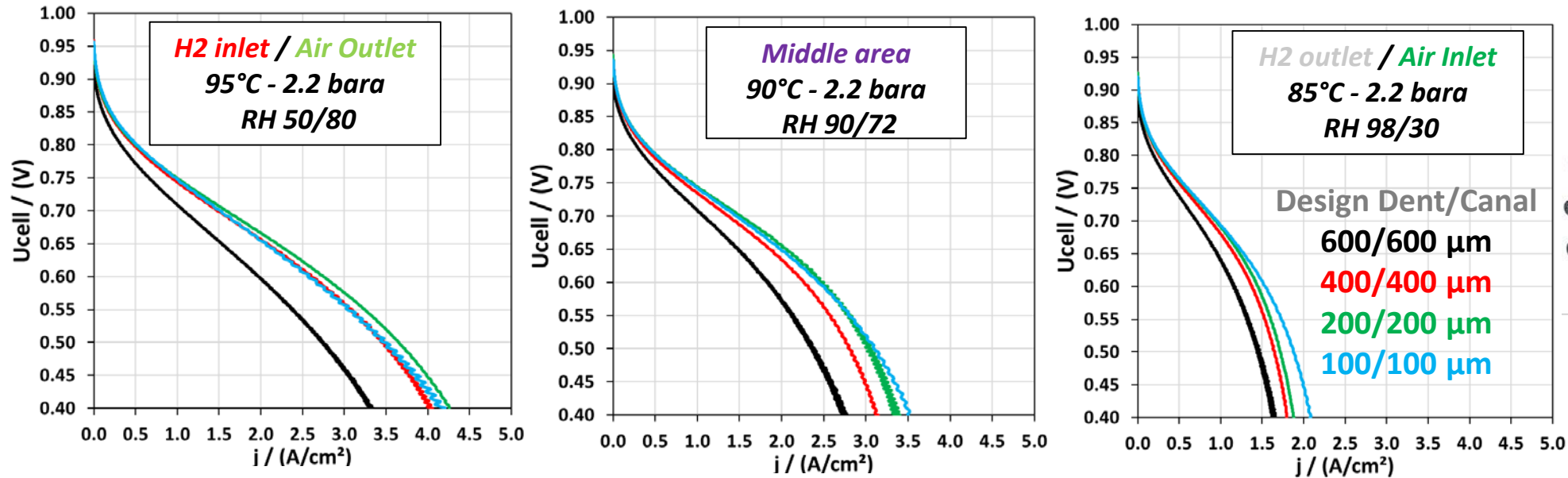


*Metrology control for
reference milled designs*



• Characterization of local performances at maximum power conditions

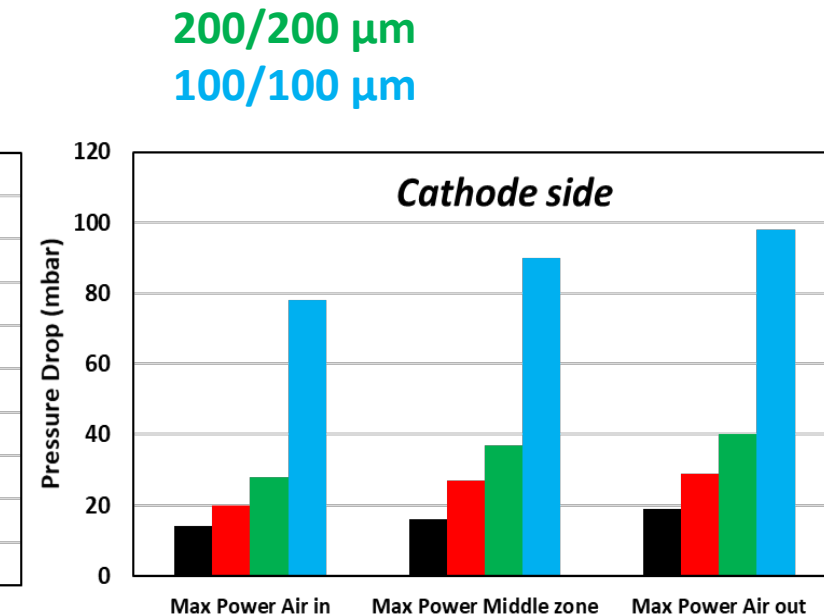
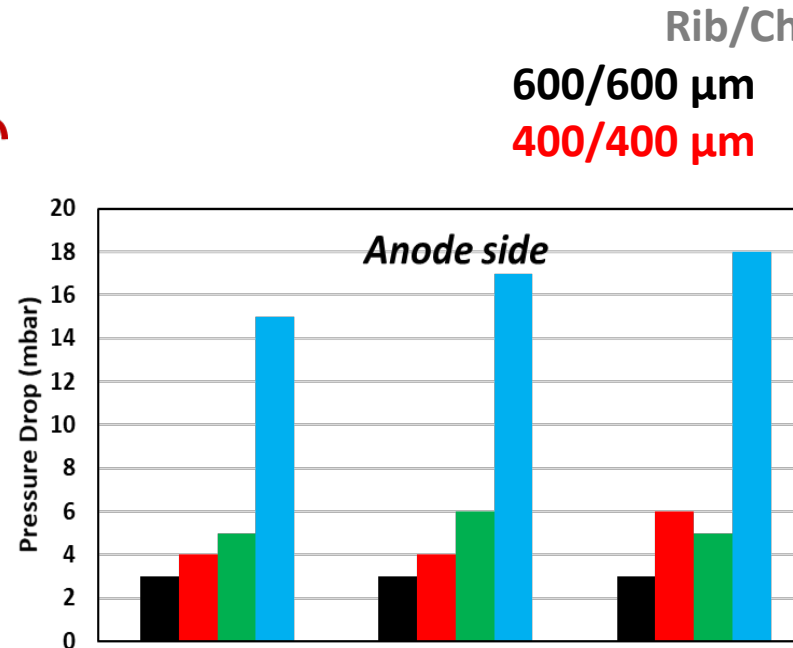
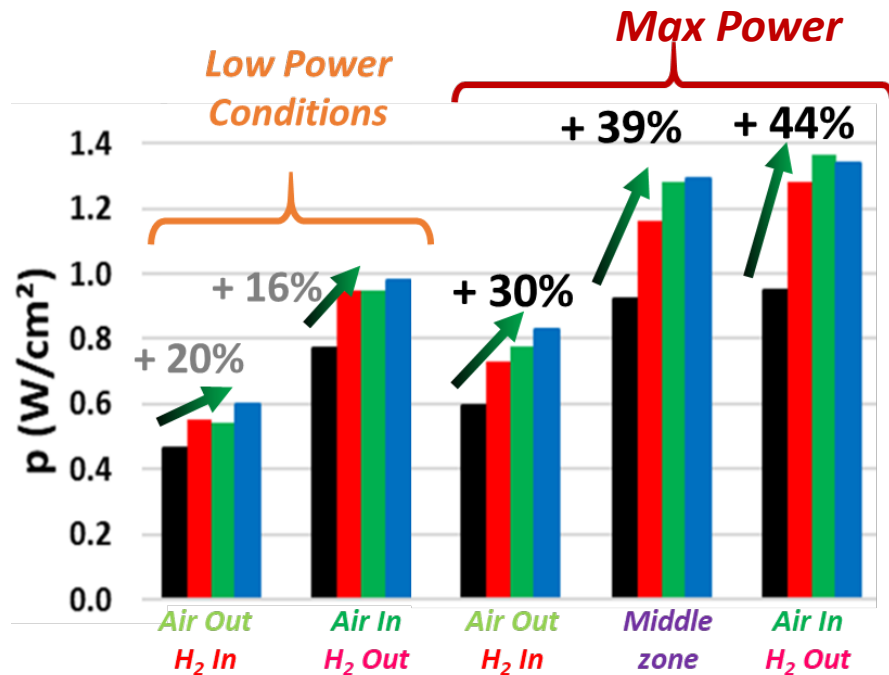
Cathode Air Flow



Anode H_2 Flow

- Performances greatly improved by reducing the rib/channel size in every local area and under every operating condition
- Even in differential/zero gradient cell with high stoichiometries : flow-field design strongly impacts the raw performances
 - High interest to decrease rib/channel size down to at least 400 μm – Minor improvement below 200 μm

Performances evaluation from local operating conditions

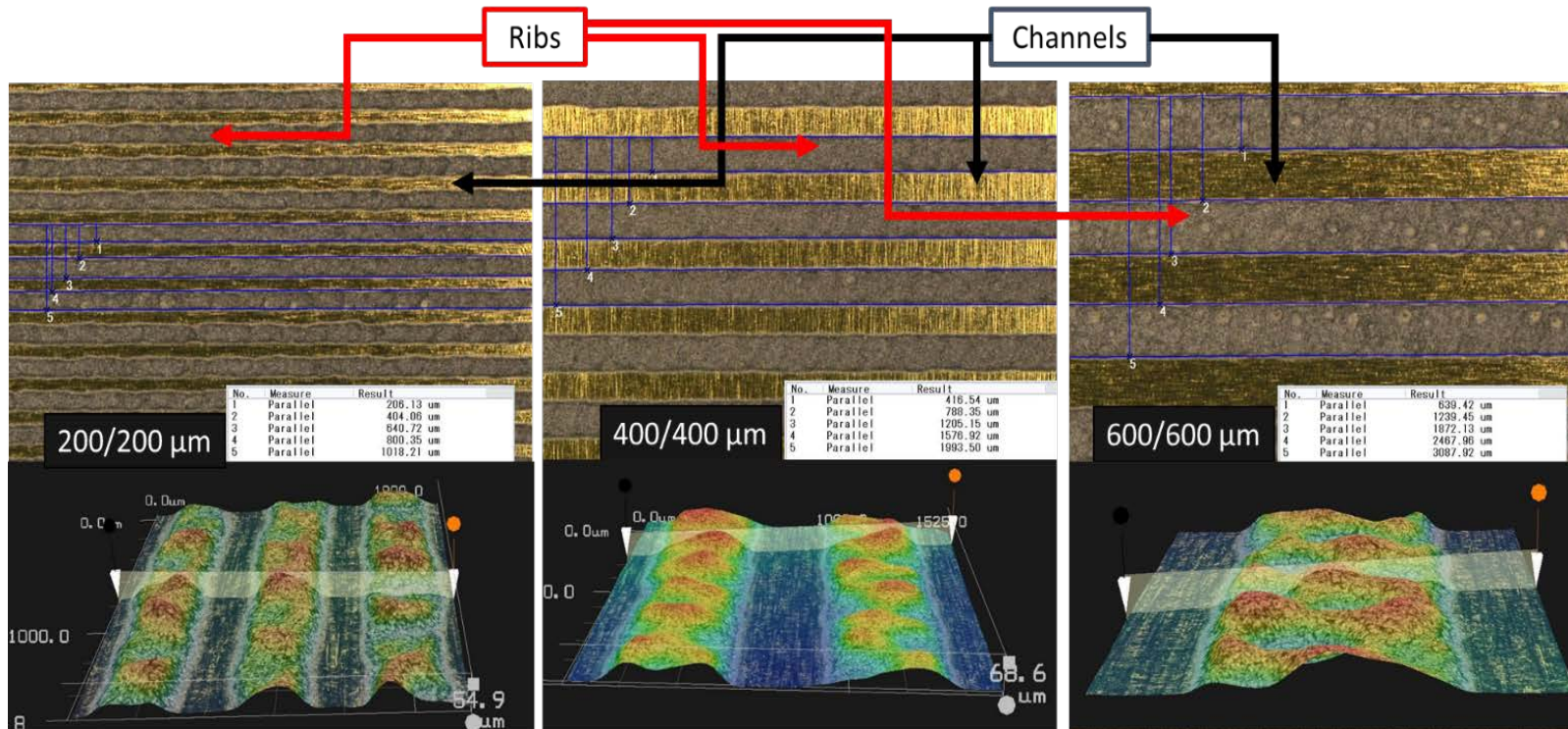


- Performance increased in every condition with refined flow field
- Power density enhancement : +30%/+44 % @ 0.66 V at max power
- No significant difference between 100/100 & 200/200 μm designs

- High pressure drop below 200/200/200 μm design
- « 100 μm size » not realistic for cathode side on large active surface area

Investigations for innovative flow-field & manufacturing processes

- Refined flow-field not achievable by SoA metallic sheet stamping
 - New manufacturing methods have to be found
 - Carbon ribs printing onto flat substrate
 - Channel integration into GDM layer
 - Additive manufacturing plates / Laser milling
- Preliminary developed/tested at small scale (2 cm²)
- Prototypes directly tested at 100 cm²

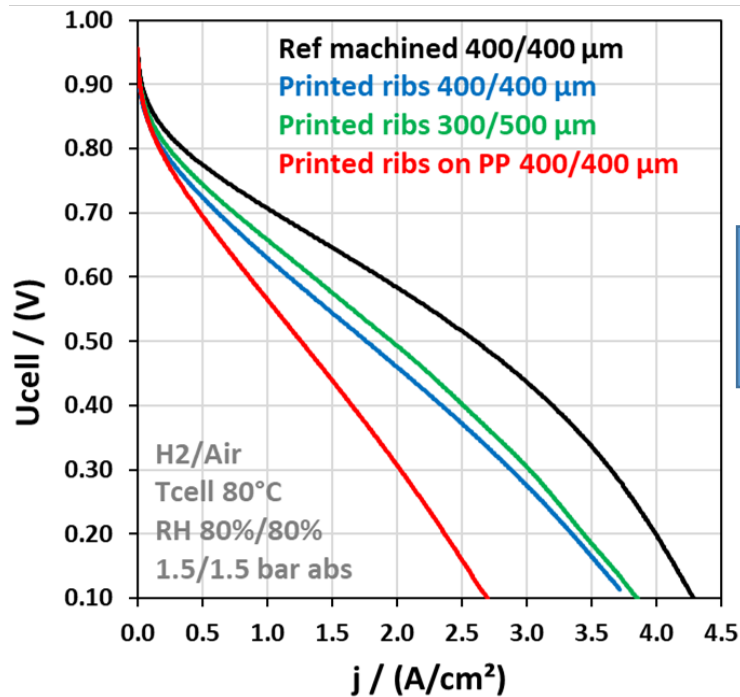


- Very thin designs can be obtained by printing carbon ribs
- Very flexible manufacturing process by screen printing

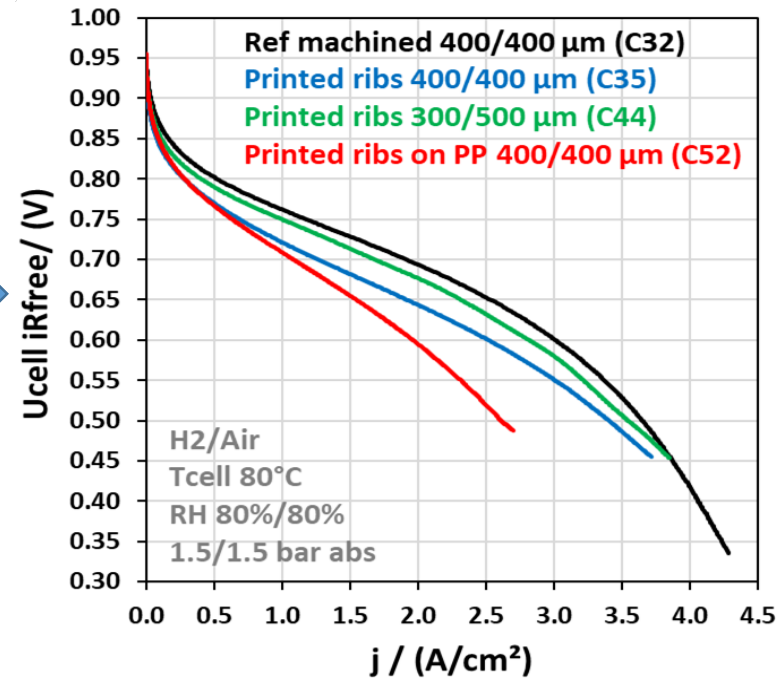
Investigations for innovative flow-field manufacturing processes

- Refined flow-field not achievable by SoA metallic sheet stamping
- New manufacturing methods have to be found
 - Carbon ribs printing onto flat substrate
 - Channel integration into GDM layer

→ Preliminary developed/tested at small scale (2 cm²)



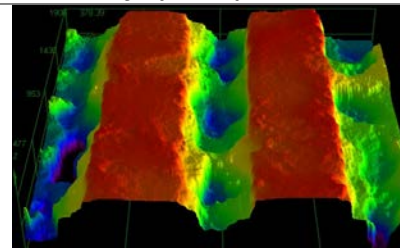
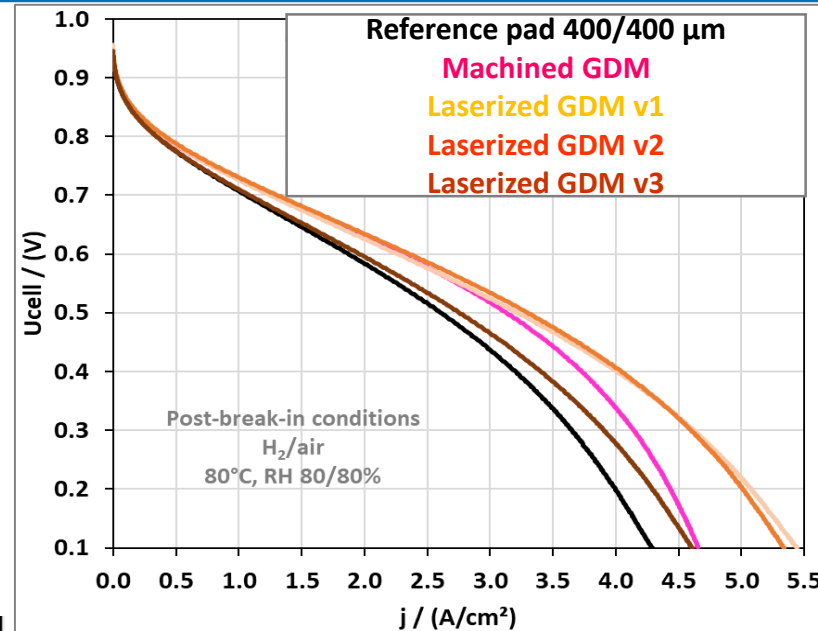
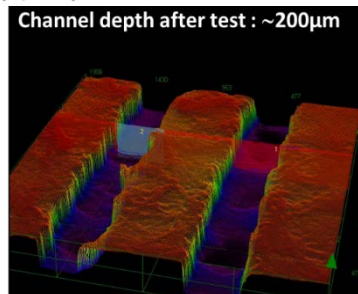
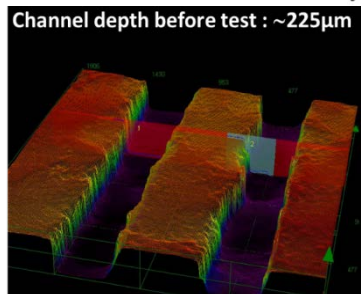
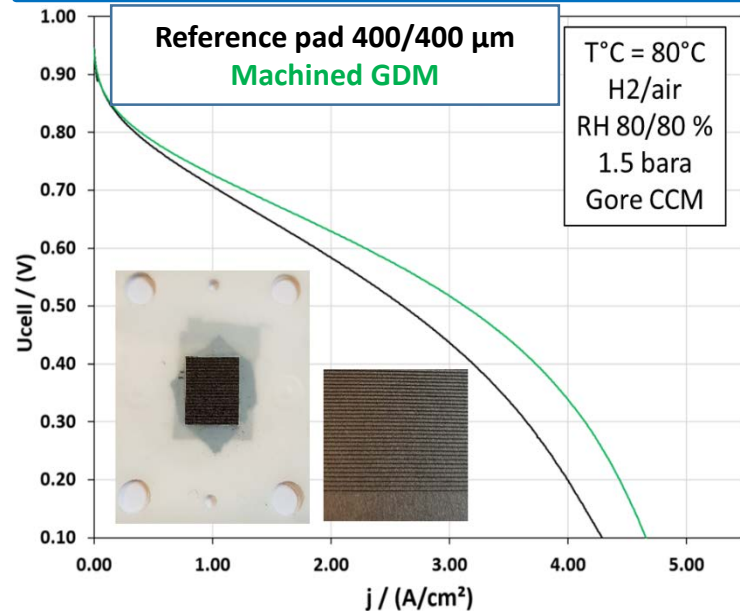
Ohmic
drop
correction



- Lower electrochemical performances
- Gap mainly due to intrinsic electronic resistance within the ribs
- Further optimization of the ink composition still in progress
- Prepreg materials : not suitable so far to be integrated as substrate

Investigations for innovative flow-field manufacturing processes

- Refined flow-field not achievable by SoA metallic sheet stamping
 - New manufacturing methods have to be developed
 - Carbon ribs printing onto flat substrate
 - Channel integration into GDM layer
- Preliminary developed/tested at small scale (2 cm²)



- Require thicker GDM/GDL to create channels (but overall cell thickness decreased) and specific GDL structure

- Alternative process tested by laser milling : Possible at small scale but further development needed at large scale (GDL deformation/twist caused during the process)

- Association of refined flow-field designs with single or dual GDM removal

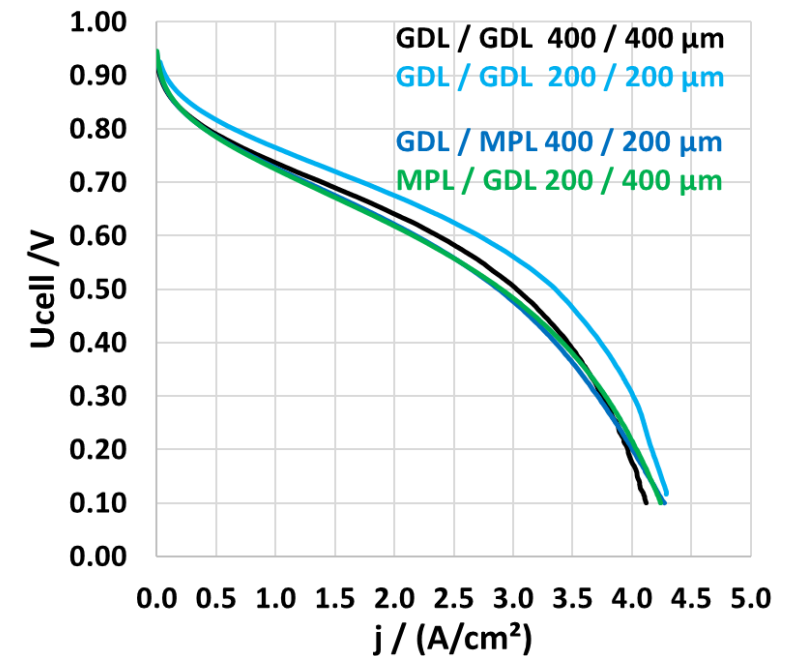
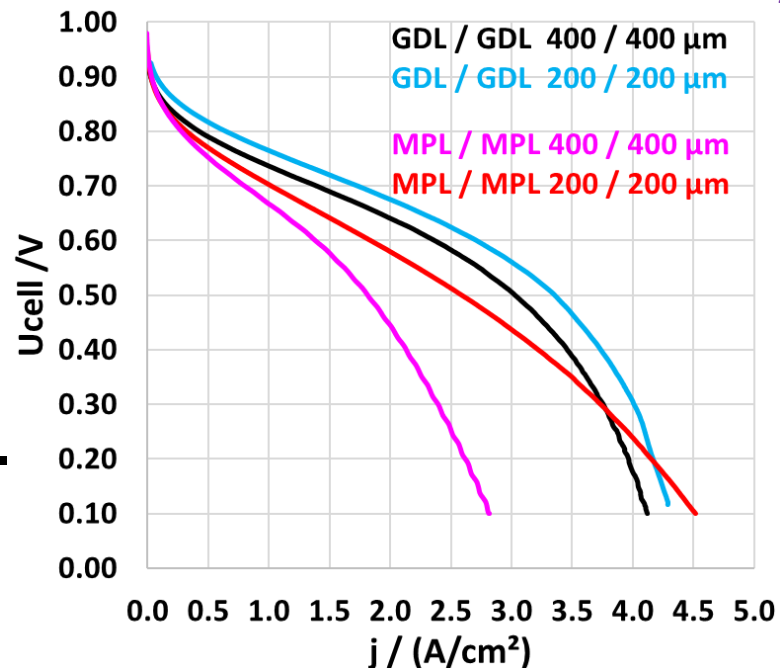
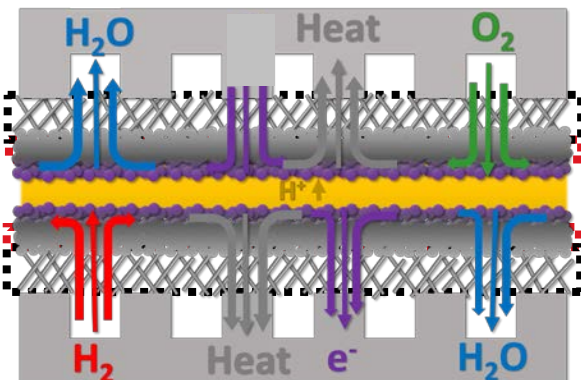
State-Of-Art Cell Architecture « GDL / GDL »

GDM layer

~ 100 - 200 μm

MPL Layer ~ 20-40 μm

EC Layer ~ 5-10 μm

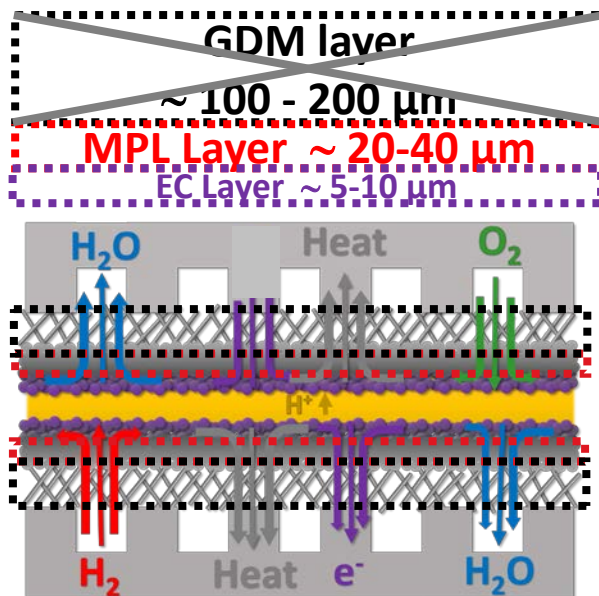


- Very poor performance without GDM with 400/400 μm design
- Higher performances with 200/200 μm but still lower than reference GDL/GDL configurations

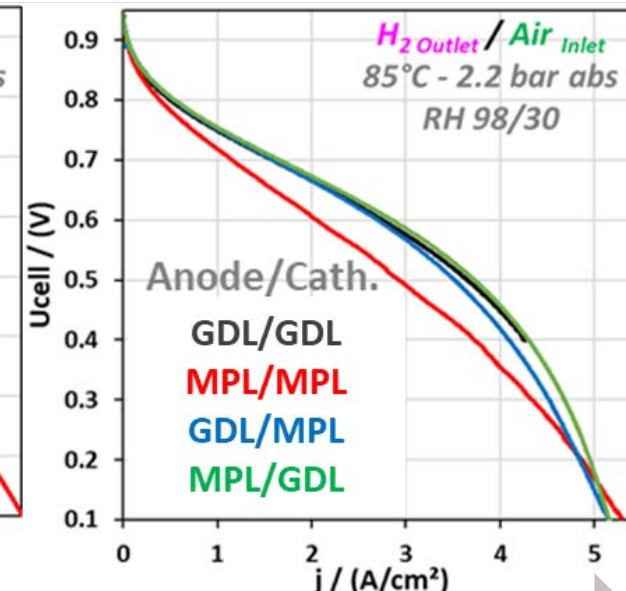
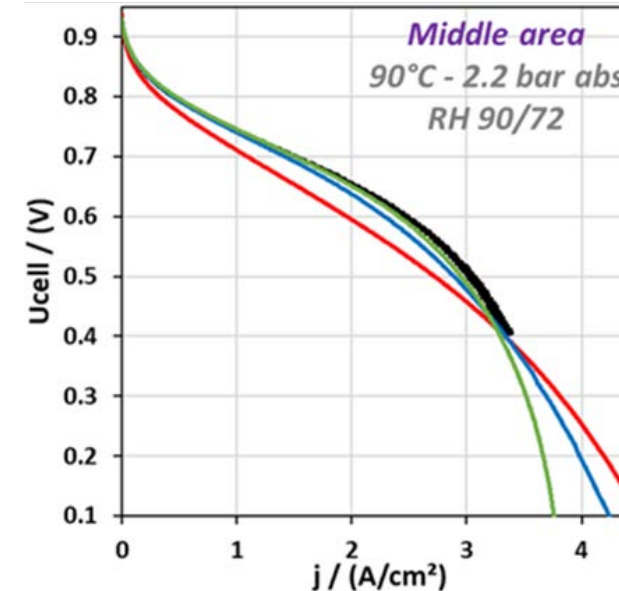
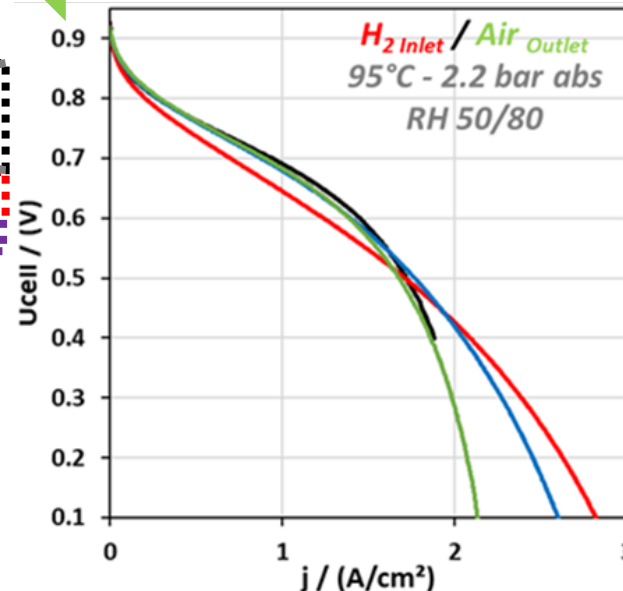
- Suppressing one GDM enable to reach satisfying performances post break-in under model operating conditions
- Preliminary validation of these innovative types of cell configuration

- R&D on new architectures coupling « Refined designs » and « single sided-GDL MEA »

New Cell Architecture « MPL / GDL »



Cathode Air Flow

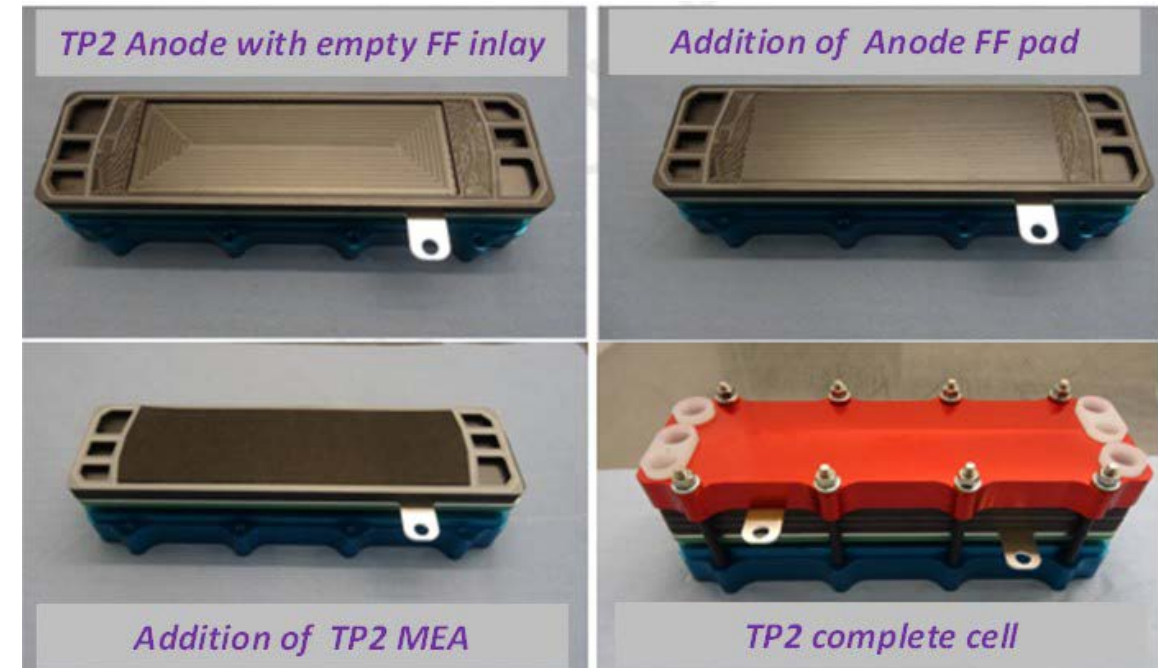
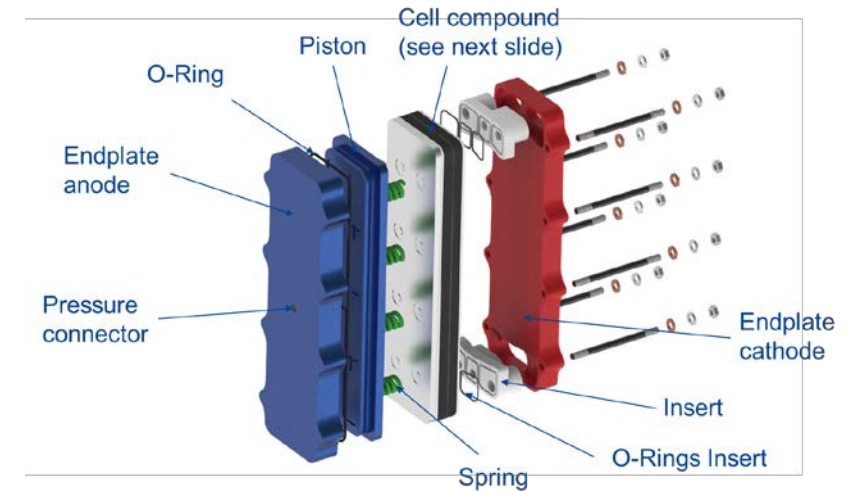


Anode H2 flow

- Both GDM suppression → Decrease of performances linked to ionomer drying at inlets/outlets and to insufficient in-plane electronic conduction within MPL
- Performances maintained or even slightly better by removing only one GDM
- Decrease of cell pitch, materials and cost saving
- Best configuration : GDL Anode + MPL on cathode active layer → Planned at 100 cm² scale

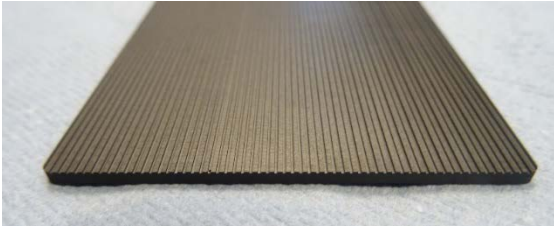
• Interest of Test Platform 2 « TP2 »

- **Scaling up for EC/EFC** to representative active area size (100 cm²)
- **Validation of cell electrochemical performances under several EU conditions from previous projects** (EU Harmonized from JRC recommendations, AutoStackCore, GAIA...)
- Characterizations of **fluidics and thermal behavior** to support stack design
- Validation of EC core formulation and **further insights for local texturing**
- **Performances/Durability characterizations** in representative and heterogeneous operating condition
- **Hardware can be reused for TP3 short-stack**

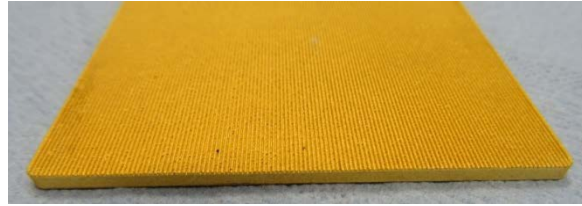


Innovative EFC: Additive manufacturing process

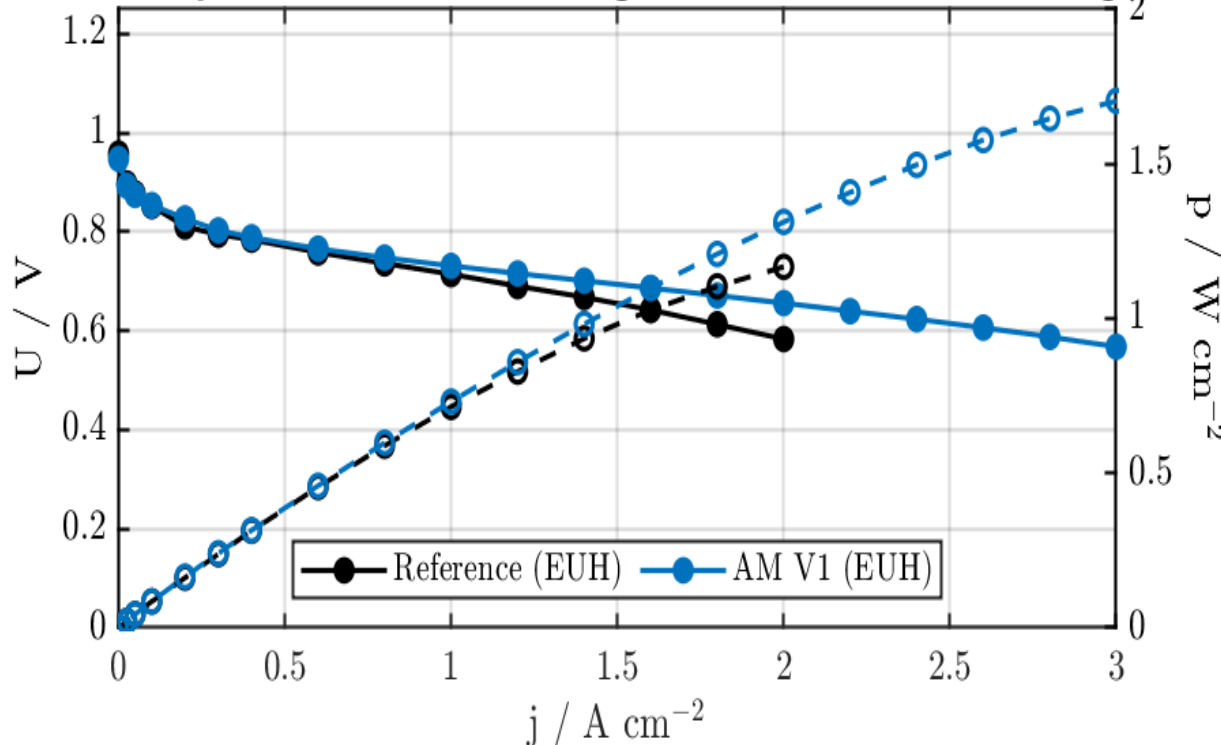
Reference Flowfield Carbon composite



AM V1 Flowfield Stainless steel + gold coating



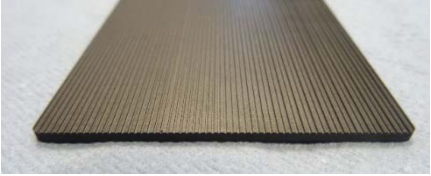
Comparison Reference Design & Additive Manufacturing



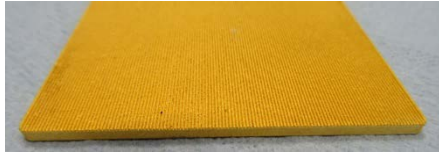
- **Reference EFC**
 - Channel/rib/depth
 - Ref Anode: 400 / 400 / 200
 - Reference Cathode: 500 / 500 / 300
- **Tests of 2 versions of AM TP2 EFC**
 - Channel/rib : Anode & Cathode
 - **AM v1 : ~ 330 / 260 μm**
 - AM v2 : ~ 300 / 200 μm
- **Use of EU-Harmonized conditions**
(pressure regulated at cell outlet)
- **Performances AM v1**
 - 3 A/cm² @ 0.58 V : 1.75 W/cm²
- **AM v1 pressure drops**
 - ~ 50 / 260 mbar @ 0.66 V (ca. 2 A cm⁻²)

Innovative EFC: Additive manufacturing and laser milling

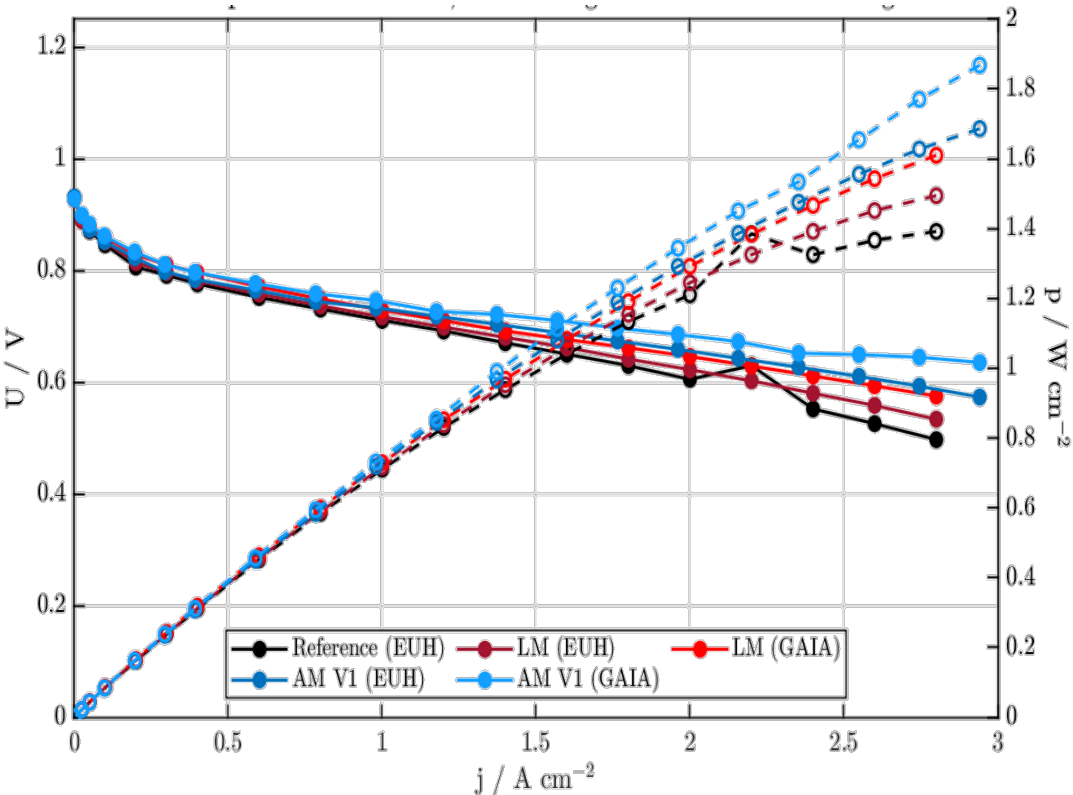
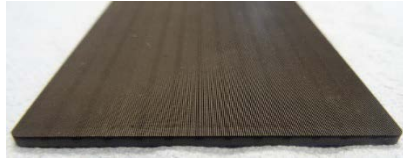
Reference Flowfield
Carbon-based



AM V1 Flowfield
Stainless steel + gold coating



Laser-Milled Flowfield
Carbon-based pad



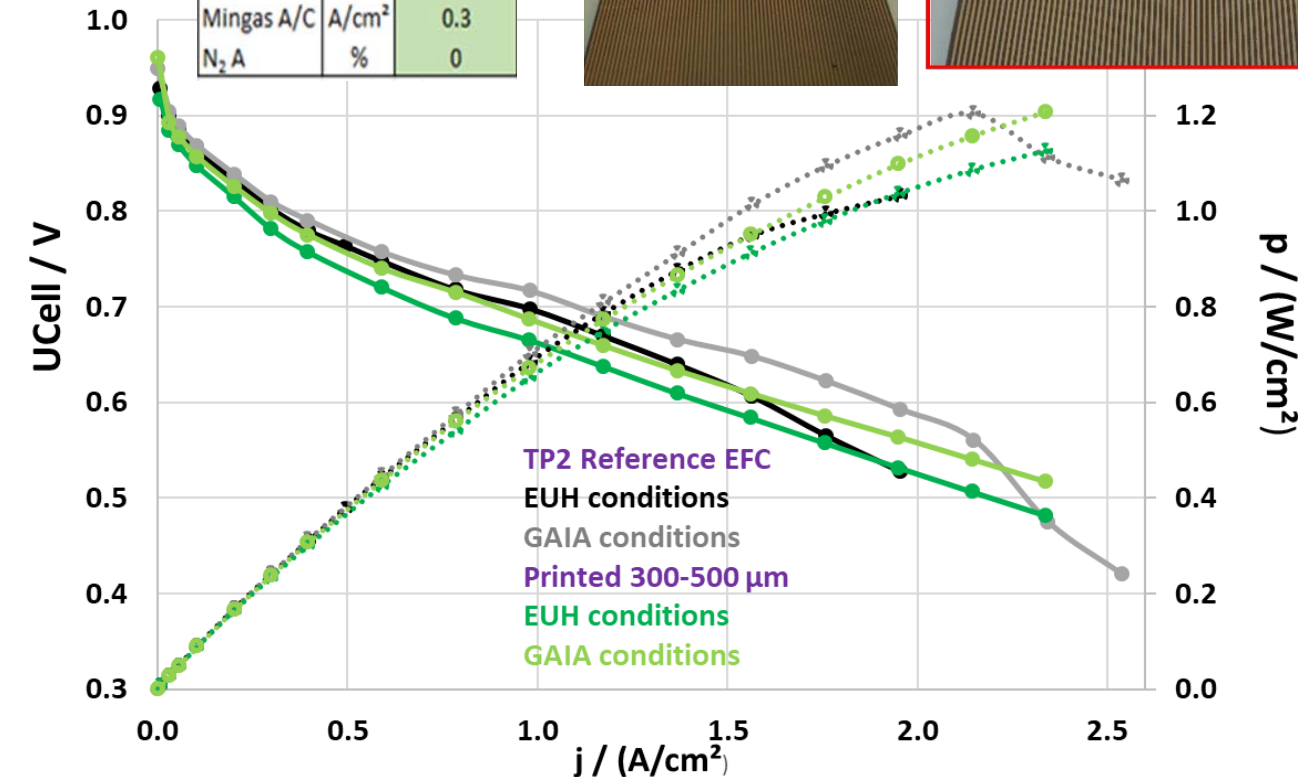
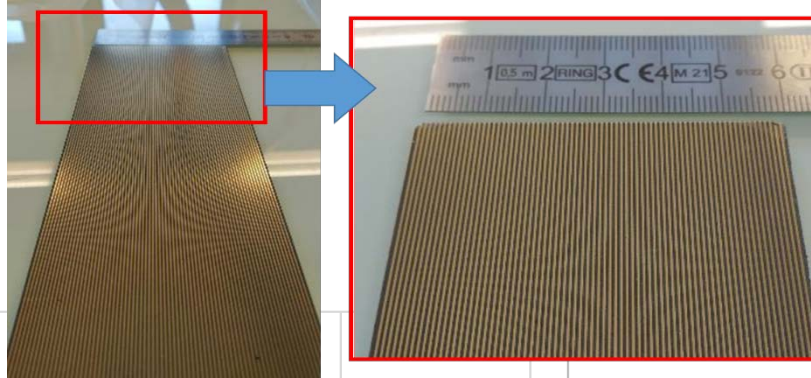
Parameter	Unit	EUH
T Cl	°C	80
T A/C	°C	82
DPT A/C	°C	64.0/53.0
RH A/C	%	50.5/30.2
Stoic A/C	1	1.4/1.6
p A/C	barg	1.5/1.3
p in/out	-	out
Mingas A/C	A/cm ²	0.3
N ₂ A	%	0

GAIA
conditions not
disclosed

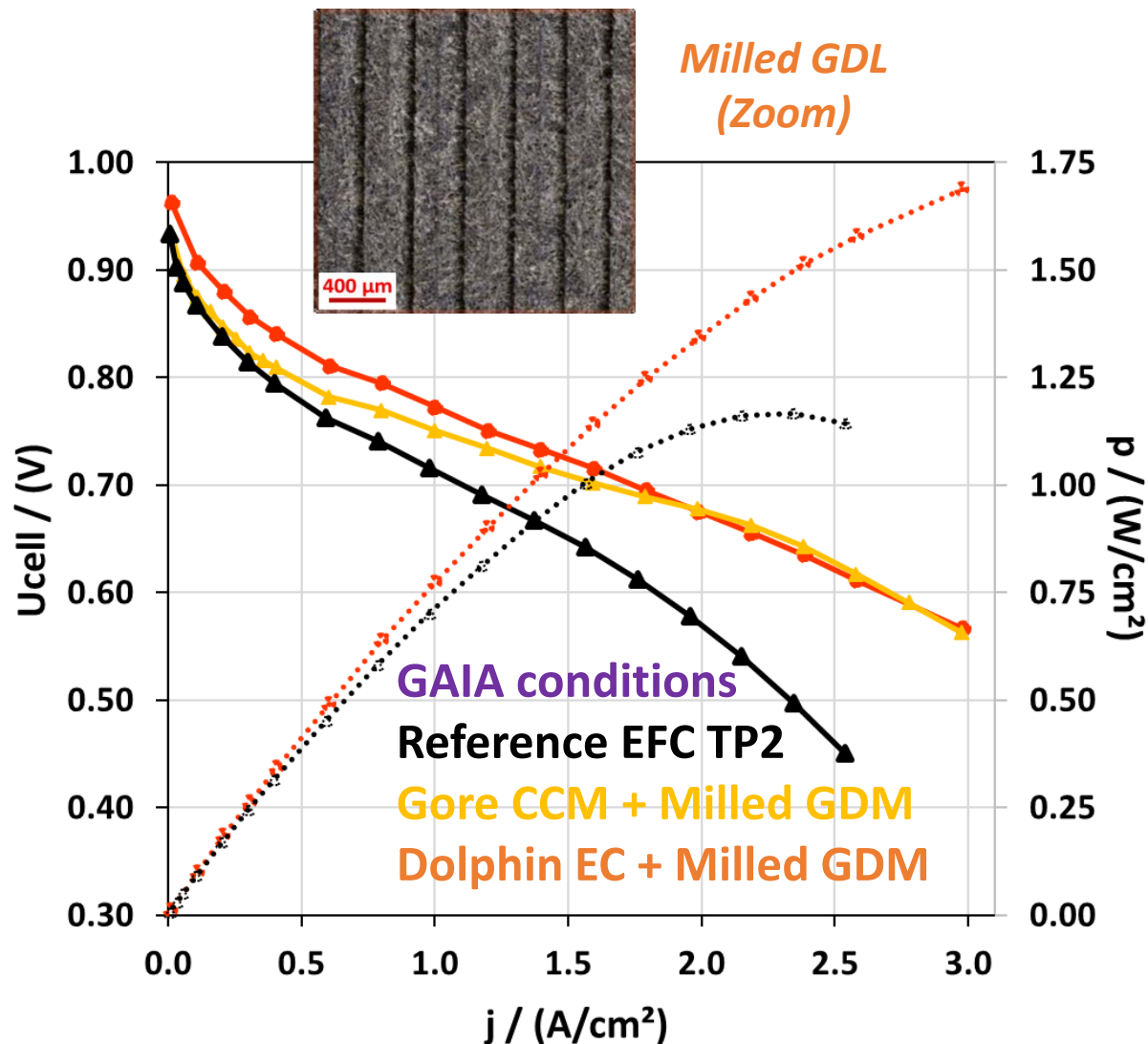
- Alternative **Laser Milled EFC**
 - Channel/rib/depth dimensions
 - Anode : ~ 200 / 100 μm
 - Cathode : ~ 200 / 100 μm
 - Very thing pattern & smaller sizes obtained for **LM** vs. **AM v1**
- Comparison between EU-Harmonized and GAIA conditions (pressure regulated at cell outlet)
- Performances **AM v1** > **LM**
- GAIA conditions leads to higher performances than EUH

TP2 pad with printed ribs

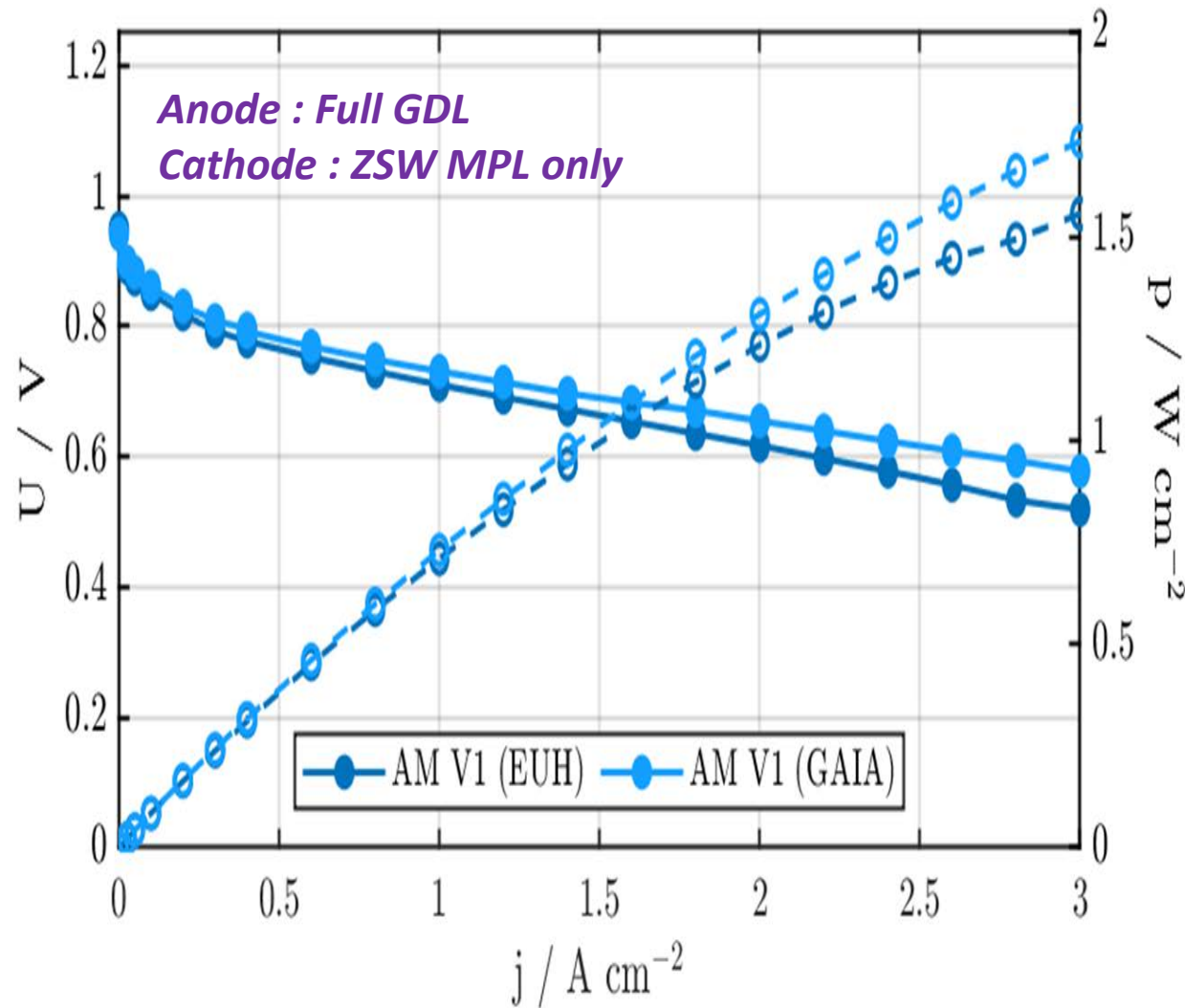
Parameter	Unit	EUH
T _{Cl}	°C	80
T _{A/C}	°C	82
DPT _{A/C}	°C	64.0/53.0
RH _{A/C}	%	50.5/30.2
Stoic _{A/C}	1	1.4/1.6
p _{A/C}	barg	1.5/1.3
p _{in/out}	-	out
Ming _{as} A/C	A/cm ²	0.3
N ₂ A	%	0



- Raw electrochemical performances slightly lower for **printed ribs** than reference design (Ohmic drop / compression effect)
- Positive effect from refined pattern for rib/channel is balanced with higher electronic resistivity within ribs.
- Technology still interesting at complete stack level in terms power density

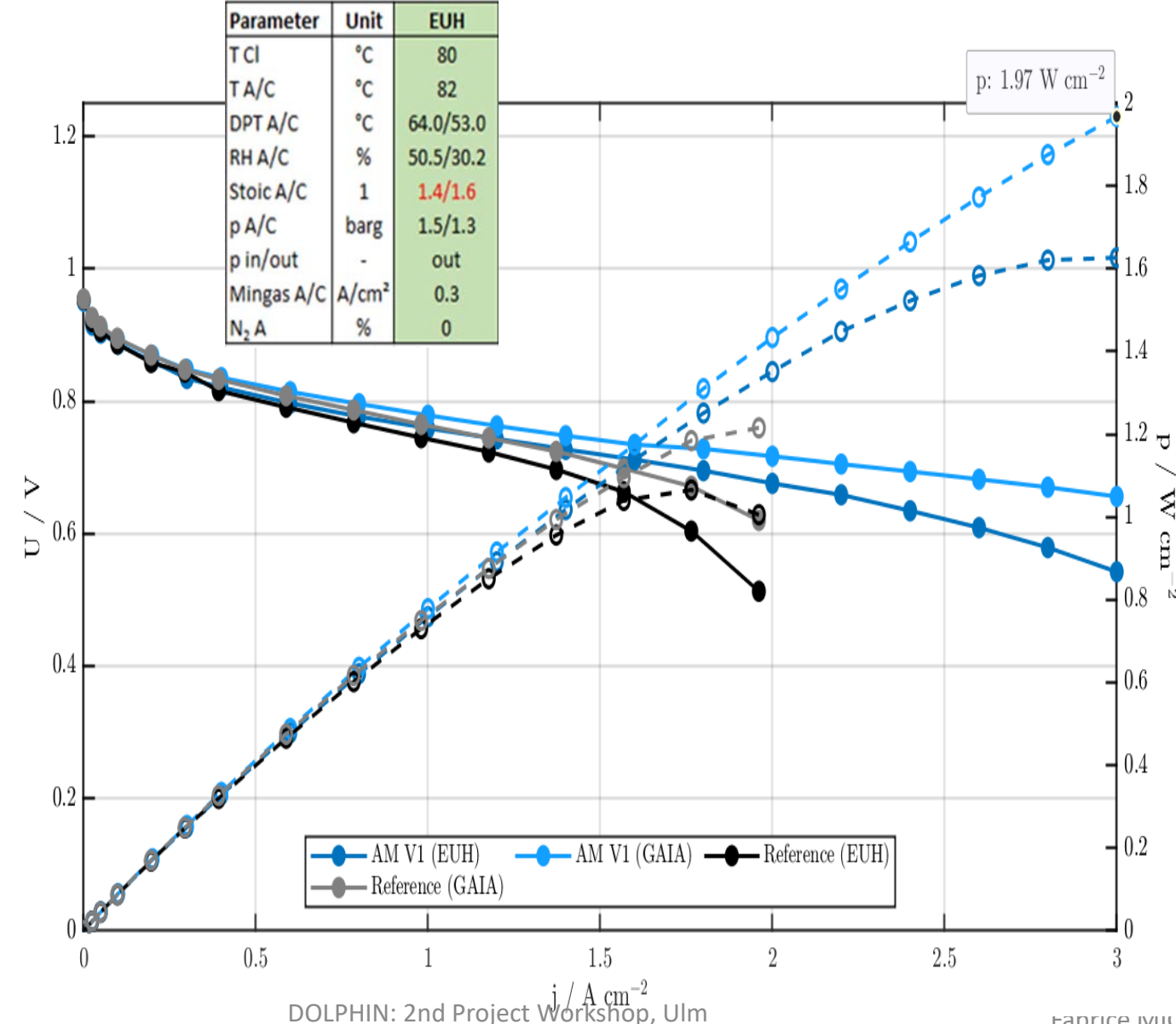


- Deposition of ZSW MPL onto machined GDM
- Cell assembly with Gore EC and advanced DOLPHIN EC
 - 2.2 A/cm^2 @ 0.66 V : 1.4 W/cm^2 ($\sim 1 \text{ W}/\text{cm}^2$)
 - 3.0 A/cm^2 @ 0.57 V : 1.7 W/cm^2
- Cell assembly and EFC concept validated at representative scale but milling not suitable with all GDM (GDL) materials in terms of thickness and mechanical properties.



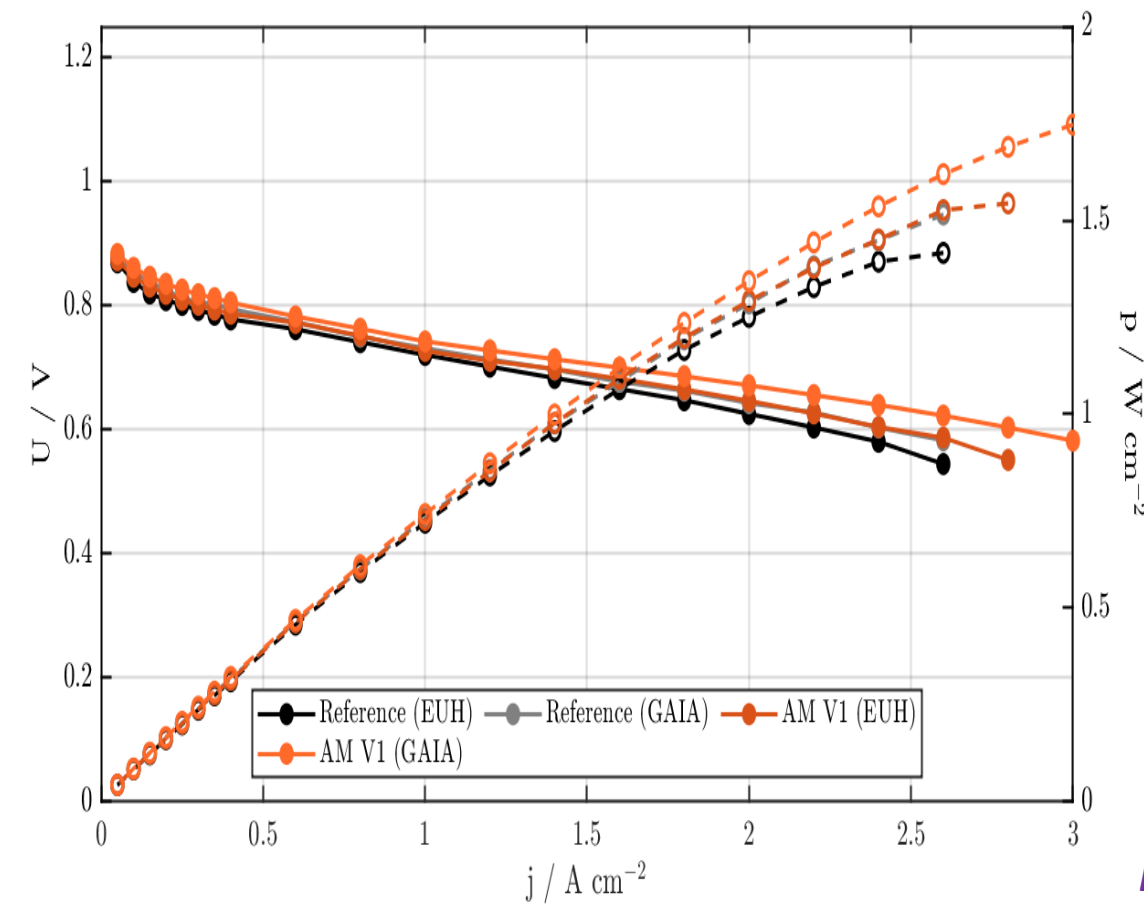
- **Tests of 2 configurations using AM v1 EFC**
 - *Single MPL Anode & GDL Cathode (not shown)*
 - **Anode GDL and MPL Cathode**
- **Single MPL at the cathode gives the best performances in 100 cm² single cell (as expected from 2 cm² single cell results)**
- **Performances obtained :**
 - **1.8 A/cm² @ 0.66 V : 1.2 W/cm²**
 - **3 A/cm² @ 0.58 V : 1.75 W/cm²**
- **Interesting Architecture regarding materials & cost saving**

TP2 : Comparison between reference EFC and EC and the best DOLPHIN concept

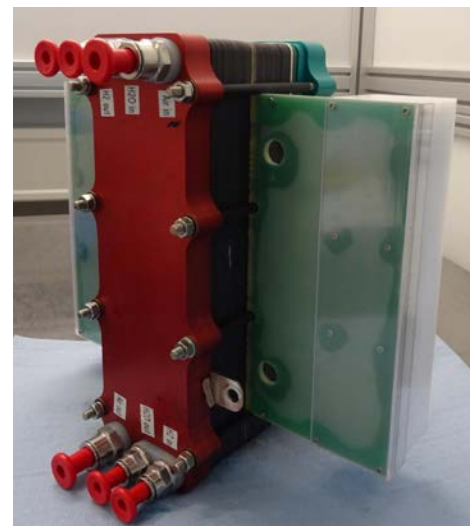
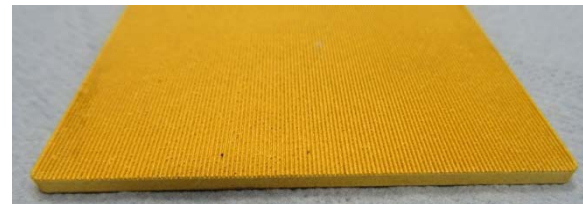


- Comparison from the reference case (SoA) and the best DOLPHIN configuration tested so far
- Reference EFC and reference EC (Gore CCM)
- AM v1 and latest DOLPHIN EC
 - RP4 DOLPHIN active layers
 - Advanced CHEMOURS membrane NDP 8011
 - Thin GDL
- Performances obtained under GAIA conditions
 - 3 A/cm² @ 0.657 V : 1.97 W/cm² !

*Polarization curves obtained in TP3
short-stack*



AM V1 Flowfield
Stainless steel + gold coating



TP3 short-stack

EFC : AM v1

EC : Gore CCM and commercial GDL

- Assembly of short-stack using **AM V1** EFC and Reference Gore EC
- **1.6 W/cm²** obtained under EUH conditions
- **1.8 W/cm²** under GAIA conditions
- Evaluation of durability in progress at ZSW using FC-DLC cycles under EUH conditions
- Performances and durability to be compared with full-stacks (Q3/Q4 2023)

Disruptive pemfc stack with n**O**vel materia**L**s, **P**rocesses,
arch**H**itecture and optimized **I**Nterfaces

DOLPHIN Workshop, Ulm June 16th 2023

Estimation of KPIs

J.-P. Poirot-Crouvezier, all



• Objective

- Estimate the gravimetric and volumetric power density for a stack with the different EFCs studied

• Results

- Gravimetric and volumetric power density for a stack
 - Ideal (at 2 W/cm²)
 - Target = 3 A/cm² @ 0,66 V
 - Expected considering experimental results
 - △ P_{nom} = performance @ 0,66 V
 - △ P_{max} = performance at maximum cell power

• TP1 (2 cm²)

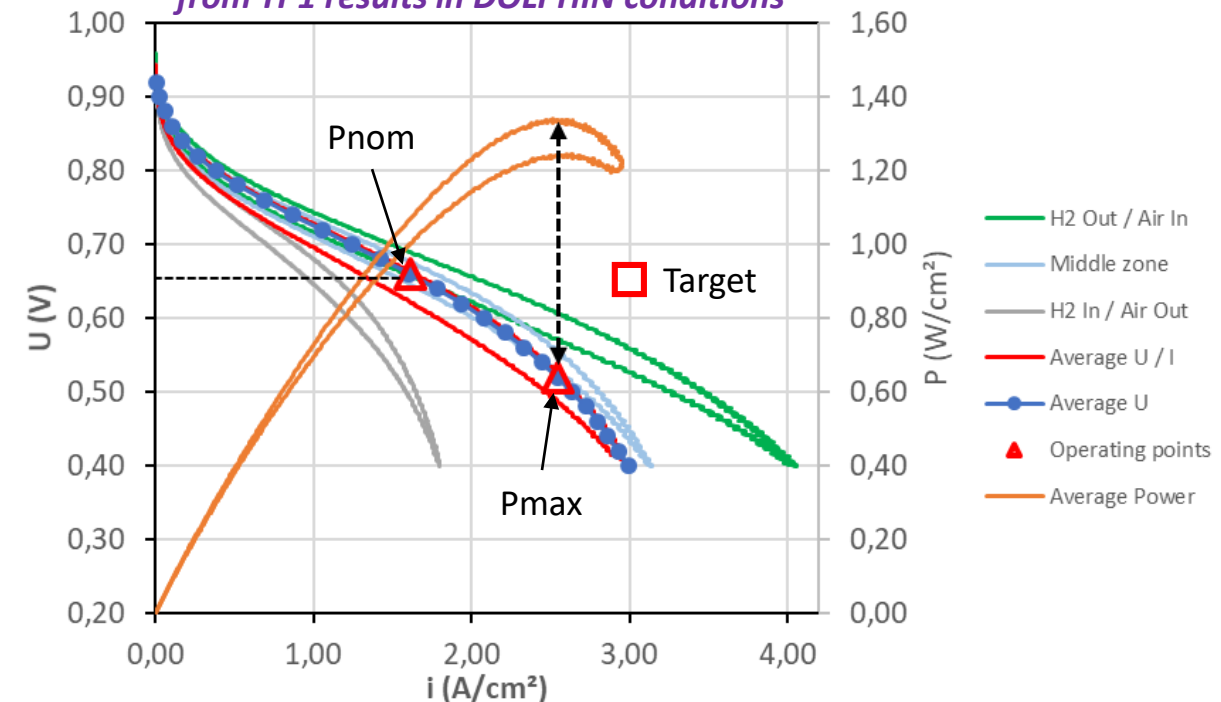
- 3 local operating conditions corresponding to cell inlet/middle/outlet for DOLPHIN « Max power » condition
 - Extrapolated cell polarization curve → P_{nom}/P_{max}

• TP2 (100 cm²)

- European Harmonized conditions (EUH)
- GAIA conditions
 - P_{nom}/P_{max} for each operating condition

Local conditions	T / °C	P A/C bar abs	RH A/C (%)	% O2 in dry gas	H2 Flow rate NI/h	Air Flow rate NI/h	N2 Flowrate NI/h
Based on max power							
H2 outlet / Air Inlet	85	2.2 / 2.2	98 / 30	21	38	95	--
Middle zone	90	2.2 / 2.2	90 / 72	14,5	38	65.6	29.4
H2 Inlet / Air Outlet	95	2.2 / 2.2	50 / 80	7,8	38	35.3	59.7

Example of polarization curve obtained from TP1 results in DOLPHIN conditions



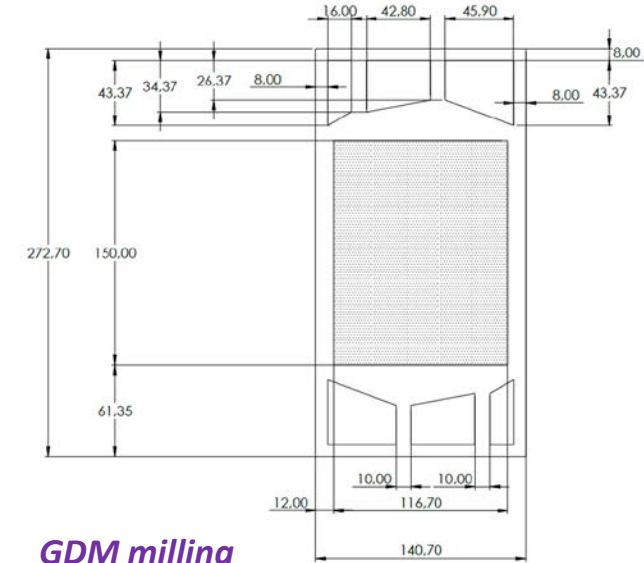
Sizing of 100 kW stacks

Input data for each EFC

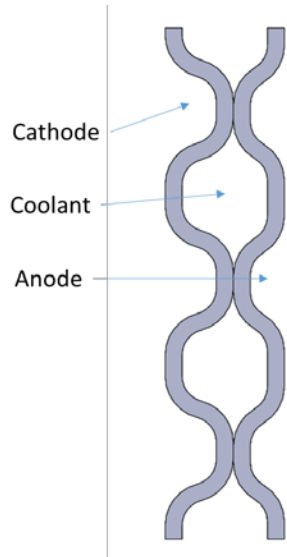
- General dimensions of TP4 cells
 - 175 cm² active area
 - 380 cm² active area
- Specific geometrical data (material density, thickness of the different layers...)

Stack hardware

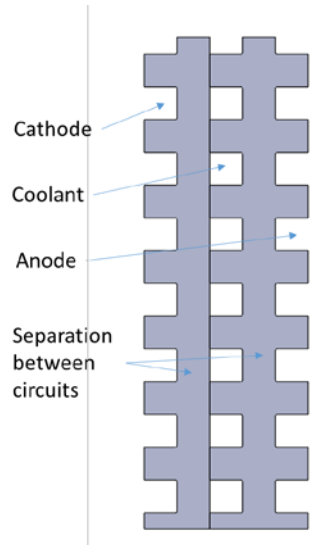
- Additional weight = 5 kg
- Additional volume = 2,5 liters
- Adjusted parameter**
 - Number of cells in the stack



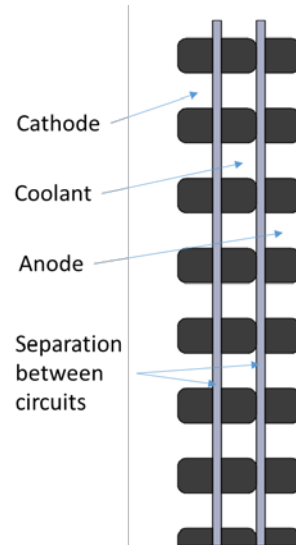
Stamping (reference)



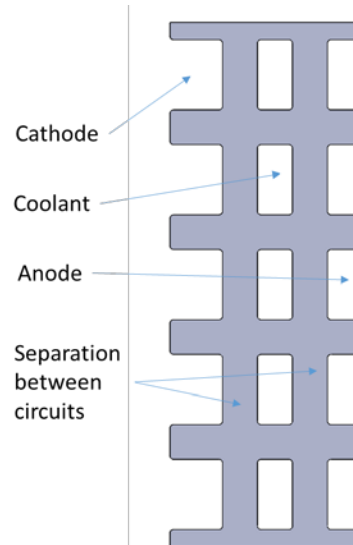
Laser Milling of carbon plates



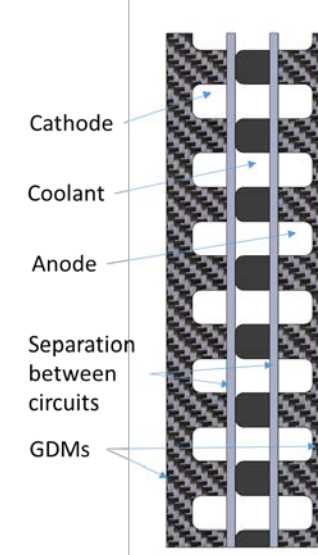
Printing on stainless steel sheets



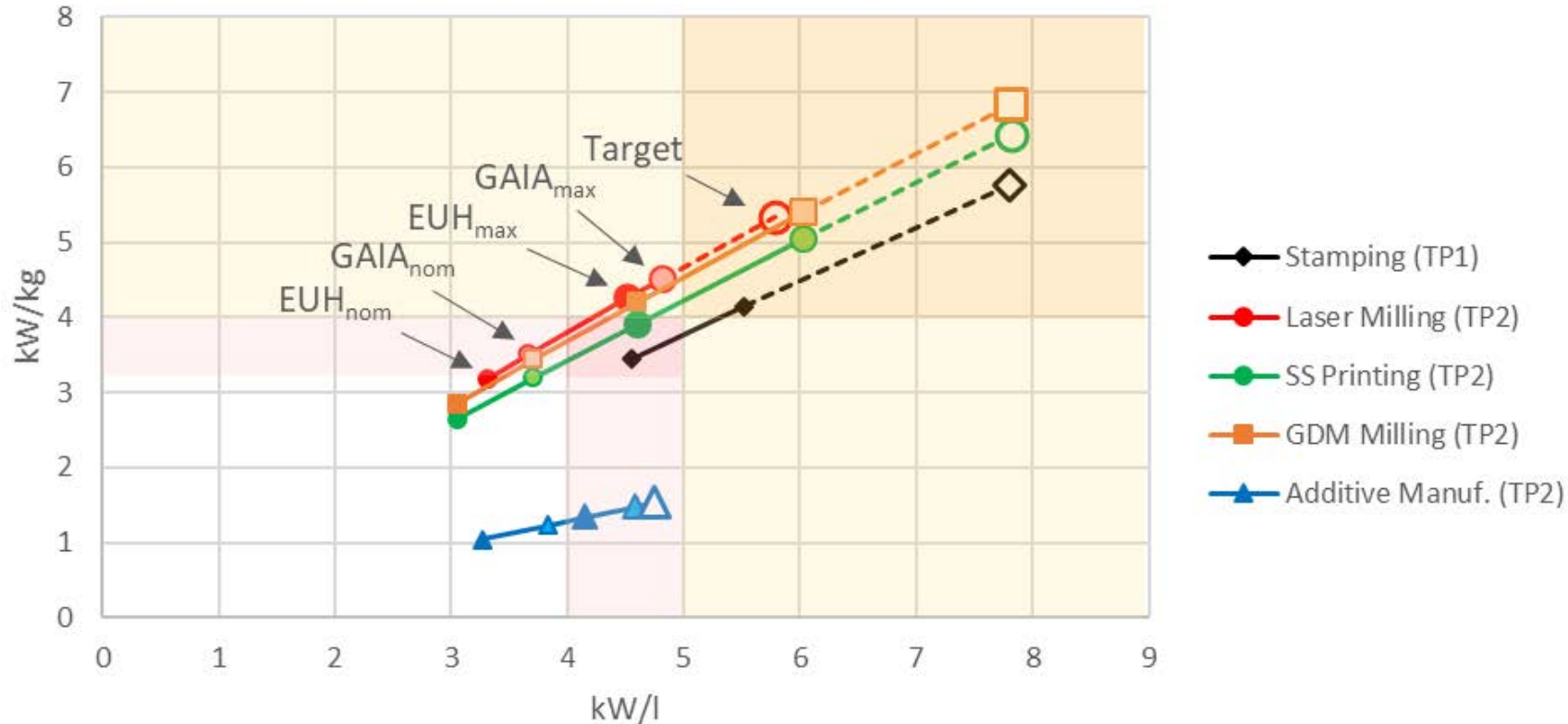
Stainless steel additive manufacturing



GDM milling



KPI values for several EFCs



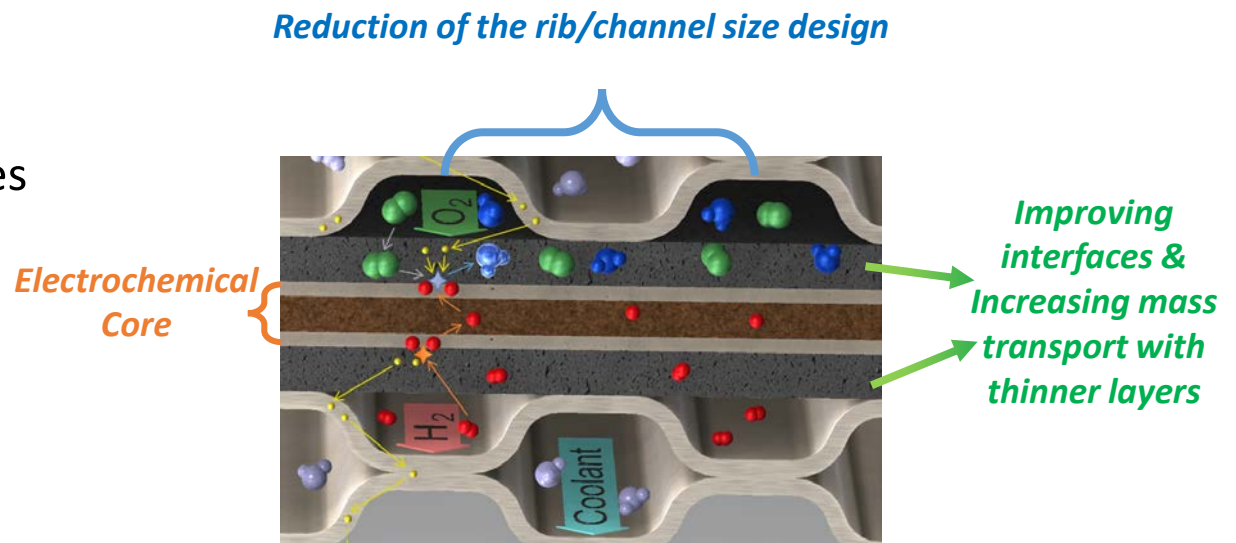
- **AM is very close to target for GAIA_{max}**
 - Gravimetric power density remains low due to the density of stainless steel
- **GDM Milling and Printing don't reach 2W/cm² but power densities are high thanks to thin metal sheets**

Disruptive pemfc stack with n**O**vel materia**L**s, **P**rocesses,
arch**H**itecture and optimized **I**Nterfaces

DOLPHIN Workshop, Ulm June 16th 2023
Back-up



- **Experimental strategy** for the validation of innovative concepts for Electrochemical and Electric and Fluidic Cores
- **Main aspects first studied at small scale**
 - **Electric and Fluidics Core**
 - Impact of the rib/channel design on the performances
 - Innovative fabrication processes
 - **Optimization of the EC|EFC interface**
 - Development of self-standing MPL materials
 - Towards the suppression of GDM support materials to reduce cell thickness
 - **Optimization of the EC**
 - Integration of new materials (membrane, catalysts)
 - Optimization of electrode composition
- **Validation of performance at representative scale**



- Characterizations of reference EC and developement in RP1 and RP2

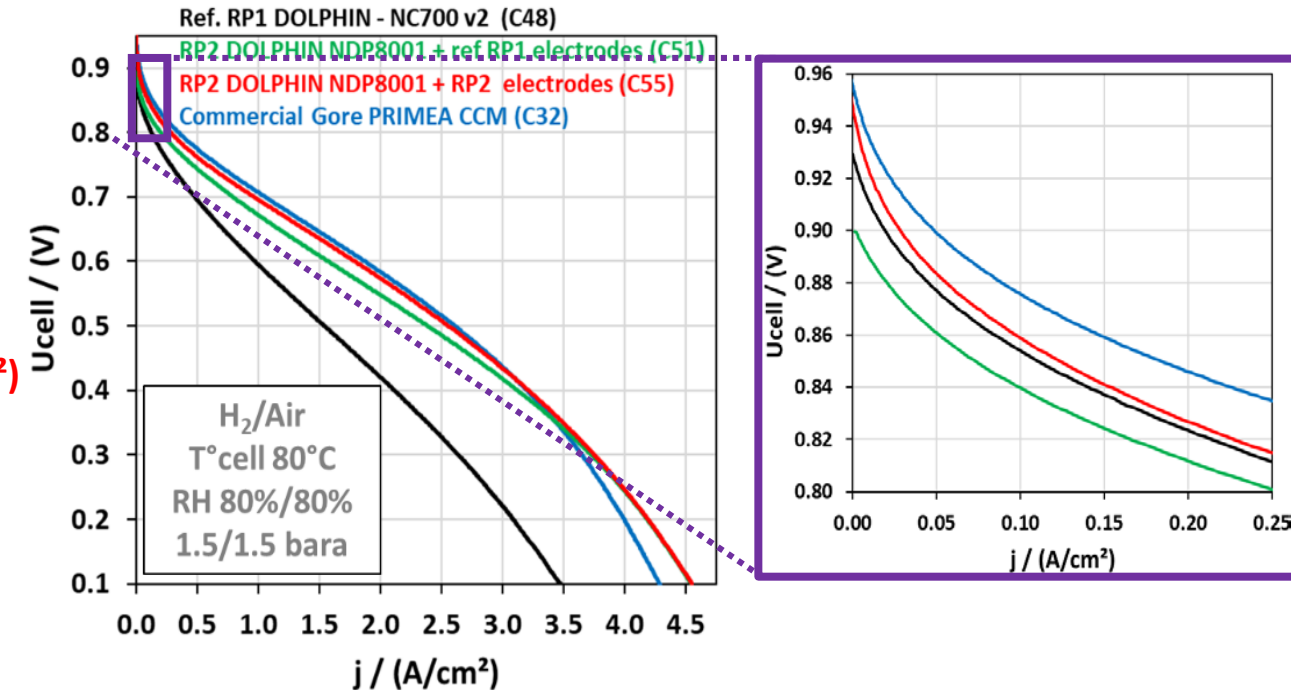
- Commercial Gore CCM (0.1/0.4 mgPt/cm²)
- DOLPHIN RP1: Ref. membrane CHEMOURS + RP1 electrodes (0.1/0.4 mgPt/cm²)
- DOLPHIN RP2 : NDP8001 + RP1 electrodes (~ 0.1/0.4 mgPt/cm²)
- DOLPHIN RP2 : NDP 8001 + RP2 electrodes (~ 0.05/0.3 mgPt/cm²)

- Integration of new membranes from CHEMOURS

- Thinner membrane: 10 μm vs 18 μm
- Higher performances / increased proton conductivity
- NDP8011 version available for the project since 2022

- Modification/Improvement of electrodes manufacturing
Softer manufacturing conditions

- Less mechanical stress for the membrane (+ 50 mV @ OCP)
- Better electrochemical activity at low overpotential...



EC type	An/Cath. Pt Loading (mg/cm ²)	@ 0.66 V		@ 0.50 V	
		j / (A/cm ²)	p / (W/cm ²)	j / (A/cm ²)	p / (W/cm ²)
Ref DOLPHIN from RP1	0.1/0.4	0.66	0.44	1.54	0.72
NDP 8001 + RP1 electrodes	0.16/0.4	1.09	0.72	2.39	1.20
NDP 8001 + RP2 electrodes	0.023/0.26	1.30	0.86	2.56	1.25
Gore PRIMEA CCM	0.1/0.4	1.39	0.92	2.61	1.30