camelot

UNDERSTANDING CHARGE, MASS AND HEAT TRANSFER IN FUEL CELLS FOR TRANSPORT APPLICATIONS

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DELIVERABLE REPORT

D5.2 – FFC VALIDATION FOR SRU GEOMETRIES USING SOA MEAS	
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NATURE OF THE DELIVERABLE		
R	*Report*	***X***
P	*Prototype*	
D	*Demonstrator*	
O	*Other*	

SUMMARY	
Keywords	*Current distribution plate, CDP, gradient MEA.* *Proton Exchange Fuel Cells (PEMFCs); Operating Conditions; Testing Protocols; Catalyst Loading Effect; Gradient catalytic loading on catalytic layer.*
Abstract	*The development of high performance and durable proton exchange membrane fuel cells (PEMFCs) increasingly relies on the components made of. One of the main components is the catalyst coated membrane (ccm). The EU-funded project Camelot aims to understand limitation of different state of the art CCMs by checking how different CCM parameters can influence the overall performance of the PEMFC. Therefore, it can help to develop a PEMFC with higher performance and durability.* *In this case study, two different full size membrane electrode assembly (MEAs) were made for a stack size PEMFC equipped with a current distrubiton plate (CDP). The two MEAs consist of membrane thickness of 10 µm, and average loading of the cathodic catalyst is 0.1 mg_{Pt} $cm^{-2}{}_{Pt}$. The main different was the gradient loading of the catalyst concept, where the loading of the CCM in the inlet side is 0.05 mg_{Pt} $cm^{-2}{}_{Pt}$ and outlet had a load of 0.15 mg_{Pt} $cm^{-2}{}_{Pt}$. The two MEA were measured in a stack PEMFC with CDP to collected and obtain a current distribution map generated by the MEAs.*
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FFC VALIDATION FOR SRU GEOMETRIES USING SOA MEAS

CONTENTS

1 INTRODUCTION

Proton exchange membrane fuel cells (PEMFC) are considered to be a good candidate for zero-emission vehicles because of their remarkable energy density, low temperature operation, and high efficiency. The EU funded project CAMELOT led by SINTEF and in collaboration with PowerCell Group, FAST Simulations, Johnson Matthey Fuel Cells, IMTEK, Chemnitz University of Technology, BMW, and PRETEXO focuses on understanding limitations of PEMFC performance of state-of-the-art (SoA) and its dependence on physical parameters of the catalyst coated membrane (CCM) such as membrane thickness and load of the catalyst.
To achieve this, the CAMELOT project has conducted diagnosis on the fundamental gas and water transport properties within the cell that limit performance of the MEAs and materials. Furthermore, the CAMELOT project has produced and tested MEAs with features that can potentially enable performance increases. Alongside the physical experiments, an open-source model was developed, to enable accurate simulation of the MEA, and validated through experimental work. This tool should enable predictions and understanding the MEA performance.

One observation made in the project is that MEAs of the same type of material, but where the physical parameters differ, have differing performances. Altering the membrane thickness has its trade-off. Thinner membranes reduce the ohmic resistance, which should improve current densities and cell activity, but it also increases the crossover of hydrogen and other reactants reducing the performance.[1] Another important parameter is the load of the catalyst, since the catalyst is considered one of the most expensive components in PEMFCs.[2] Reducing the load will result in cheaper PEMFCs, but the performance of the cell will be reduced compared to PEMFCs with higher catalyst loads. It is necessary to find the optimal trade-off point to gain optimum performance versus a more affordable energy price of kW per cost. Physical component parameters will strongly affect the cell performance, cost, and efficiency.

To gain improved performance of PEMFC, it is important to understand the behaviour of PEMFC during operation when the fuel and air are flowing through the channels. As the gas flows through the channels during operation, the fuel and air are consumed, and their concentration are lower, which will influence the mass transfer and performance. Therefore, to gain better durability and performance with minimum loading of the catalyst, and to keep the production cost of the PEMFC reasonable cost is to distribute the catalyst loading in a gradient way over the CCM. With a normal non-gradient MEA will the a gradient decline performance across the chanel due to consumption for the gases. However, with gradient MEA, if loading of the catalyst distributed right, it can compensate issues or degradation that may occur.[3] For example, in theory increasing loading from low load in inlet to high load in outlet can assist with the mass transfer issue since the lower concentration gas has higher probability to encounter a catalyst due to increasing amount of loading.

This delivery package work present the study of the two concept full size MEA that were tested in a full size short stack PEMFC. The two full size MEA are regular MEA with homogenous catalytic loading, whereas the second MEA is based on gradient catalytic loading when the inlet has low loading in the air inlet and higher loading in air outlet. Those MEAs were studied by equipping the PEMFC with current distribution plate (CDP).[4] The CDP consist of matrix array of current reads sensors across the CDP, which read the current value that is in contact in the particular position on the MEA CCM. Such tool can help to understand the behaviour of the MEA during operation time, which can show the advantage and disadvantage of gradient MEA.

2 EXPERIMENTAL

Membrane electrode assemble:

Four types of prototypes MEAs were prepared and provided by Johnson Matthey Fuel Cells were tested in a short-stack full size PEMFC. The size of the CCM of the MEAs were 240 cm², where each MEA had different membrane thickness or cathode catalytic loading. Three of MEAs had regular homogenous CCM loading, while the fourth one

was based on gradient CCM loading. Table X shows the difference between the loading and membrane thickness. The gradient MEA the average loading is similar to MEA type #3, but the distribution of the loading on cathode is different across the CCM, the air inlet region was 0.05 $mg_{Pt}\ cm^{-2}{}_{Pt}$ at the inlet and 0.15 $mg_{Pt}\ cm^{-2}{}_{Pt}$ at the outlet. It is important to note that all MEAs had a minor shift with the CCM, where the sub-gasket has covered 1 cm of the inlet region part of the CCM.

Table 1, Parameters of the MEAs in this work

MEA type # MEA #1	Average cathode loading $mg_{Pt}\ cm^{-2}{}_{Pt}$ 0.4	Membrane thickness μm 15	Type CCM Homogenous
MEA #2	0.1	15	Homogenous
MEA #3	0.1	10	Homogenous
MEA #4	0.1	10	Gradient

The short stack PEMFC builds.

Two different PEMFC builds run were done in this work, where each time the PEMFC was equipped with CDP. Due to the small quantity of each prototype MEA type, the short stack PEMFC had to be consist of segments 3 different MEAs to fill a PEMFC with 10 cells. Each build the CDP was in contact with the relevant tested MEA #3 or MEA #4.

The electrochemical procedure.

Each of the PEMFC build conduct two type of polarisation curve runs, which were conducted at 70 °C and 80 °C, table 2 and 3 presenting the operation condition at respectively temperatures.

Table 2: Operating Condition at 70 °C

	Parameters	**Symbol**	**Unit**	**Values**
Coolant	Coolant Inlet Temperature	T_{cell}	°C	70
ANODE	Inlet Fuel Pressure	P_{anode}	bara	1,4 - 2,4 (varying over load)
	Inlet Fuel Stoichiometry	λ_{H2}	-	1,5
	Inlet Fuel Temperature	T_{anode}	°C	70
	Inlet Fuel Dew Point (Relative Humidity**)	T^{Dew}_{anode} (RH)	°C (%)	54 (60)
	Inlet Fuel Composition*	X_{H2} (-)	- (-)	1
C	Inlet Oxidant Pressure	$P_{cathode}$	bara	1,2 - 2,2 (varying over load)
	Inlet Oxidant Stoichiometry	λ_{O2}	-	1,8

	Inlet Oxidant Temperature	$T_{cathode}$	°C	70
	Inlet Oxidant Dew Point (Relative Humidity**)	$T_{cathode}^{Dew}$ (RH)	°C (%)	54 (60)
	Inlet Oxidant Composition*	X_{O2}	-	0,21 (Air)

*Composition given in mole fraction, dry-basis

**Relative Humidity based on Coolant Inlet Temperature

Table 3: Operating Condition at 80 °C.

Parameters		Symbol	Unit	Values
Coolant	Coolant Inlet Temperature	T_{cell}	°C	80
ANODE	Inlet Fuel Pressure	P_{anode}	bara	1,4 – 2,4 (varying over load)
	Inlet Fuel Stoichiometry	λ_{H2}	-	1,5
	Inlet Fuel Temperature	T_{anode}	°C	85
	Inlet Fuel Dew Point (Relative Humidity**)	T_{anode}^{Dew} (RH)	°C (%)	48,2 (45)
	Inlet Fuel Composition*	X_{H2} (-)	- (-)	1
CATHODE	Inlet Oxidant Pressure	$P_{cathode}$	bara	1,2 – 2,0 (varying over load)
	Inlet Oxidant Stoichiometry	λ_{O2}	-	1,8
	Inlet Oxidant Temperature	$T_{cathode}$	°C	85
	Inlet Oxidant Dew Point (Relative Humidity**)	$T_{cathode}^{Dew}$ (RH)	°C (%)	48,2 (45)
	Inlet Oxidant Composition*	X_{O2}	-	0,21 (Air)

*Composition given in mole fraction, dry-basis

**Relative Humidity based on Coolant Inlet Temperature

3 RESULTS AND DISCUSSION

Two short-stack PEMFC platforms were constructed with several different full-size MEAs. The MEAs consisted of different catalyst loadings and membrane thicknesses as a reference performance, where two different MEA concepts were tested in a short-stack PEMFC system, where the MEAs under investigation were facing the CDP of each platform. The tested MEAs had similar physical parameters, with an average load of 0.1 mgPt cm^{-2} and a membrane thickness of 10 µm. The main difference between the two types of MEAs was that one had a homogenous catalyst coating, and the other had a gradient loading of the catalyst. The loading in the gradient MEA was linear with 0.05 $mg_{Pt}\ cm^{-2}{}_{Pt}$ at the inlet and 0.15 $mg_{Pt}\ cm^{-2}{}_{Pt}$ at the outlet. The concept MEAs were in contact with the CDP to read the distribution of the current density.

Regular MEA

Two different polarisation curves were conducted at different temperatures to observe the current distribution and how it the temperature influenced the overall cell performance. The first platform build consisted of three different types of non-gradient MEAs, with each having different PEM thickness or catalyst loading, to evaluate the influence of each parameter. Table 4 presents the PEM thickness and the cathodic catalytic loading parameters of the three different MEAs tested in the stack. MEA type #3 was in contact with the CDP.

Table 4, Parameters of the MEAs that were in the first short-stack PEMFC build.

MEA type #	Average cathode loading $mg_{Pt}\ cm^{-2}{}_{Pt}$	Membrane thickness μm	Type CCM
MEA #1	0.4	15	Homogenous
MEA #2	0.1	15	Homogenous
MEA #3	0.1	10	Homogenous

The MEAs performances were tested at two different temperatures, 70 °C and 80 °C, to observe the differences. Figure 1 shows the polarisation curves of the three tested MEA inside the stack PEMFC build. The high load MEA #1 showed a better performance compared to the low load MEA #2 and #3.[5] Although MEA #2 and #3 has similar catalytic loadings, MEA #2 shows slightly better performance compared to the thinner membrane MEA #3. The lower performance could be caused by the cross-over of H2 and other reactants. The temperature had a negligible influence on the performances.

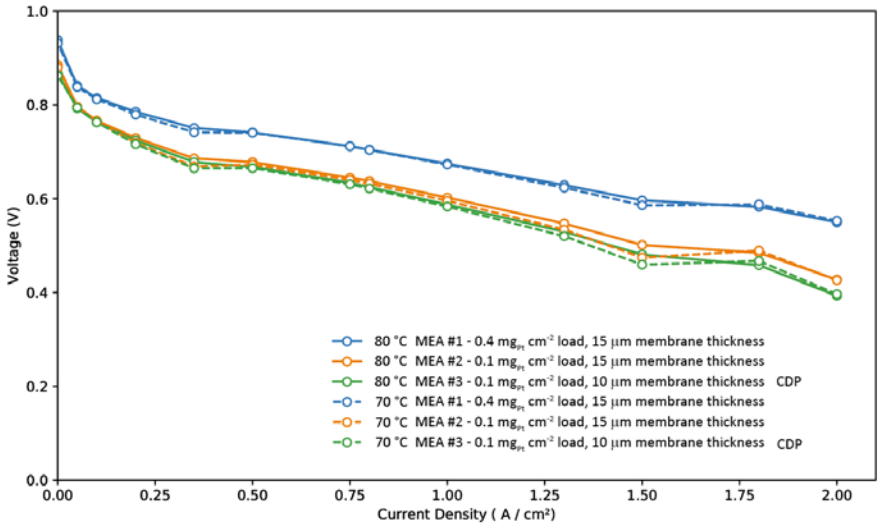

Figure 1 – The polarisation curves of three different types of MEAs that were tested simultaneously in the PEMFC stack. MEA #3, with a low loading of 0.1 mg$_{Pt}$ cm^{-2}, which was placed so that it was in contact with the current distribution plate (CDP).

Figure 2 shows snapshots of the current distribution in 2D and 3D collected by CDP in contact with MEA #3. The Figure shows snapshots of the typical behaviour of the MEA at different average current densities of the polarisation curves, for 70°C (Figure 2a) and 80°C (Figure 2b). The left side of the 2d current distribution maps show the current distribution near the H2 inlet and air outlet of the fuel cell stack, while the right side show the current distribution near the air inlet and H2 outlet. The right side of the CCM, as seen in the 2D CD maps, was slightly blocked by the subgasket, hence the activity of the first 1-2 cm were reduced.

The MEA performance at both temperatures shows similar trends in current distribution, see Figure 1. The first row in Figure X shows the activity at high current densities, i.e. in the mass transfer region. The highest current densities are located near the air inlet, where the concentration of air is naturally the highest. As the air is depleted throughout the MEA towards the air outlet resulting in lower current densities due to limited mass transfer. At a load point 1.5 A cm^{-2}, the current distribution is a gradient across the catalytic layer. At the low-mid ohmic region of 1.0 A cm^{-2},the distribution of the current is quite homogeneous except near the edges of the active area. In the kinetic region kinetic , at, 0.2 A cm^{-2}, the current distribution is almost completely homogeneous except at the right side at inlet/outlet of H2/air location.

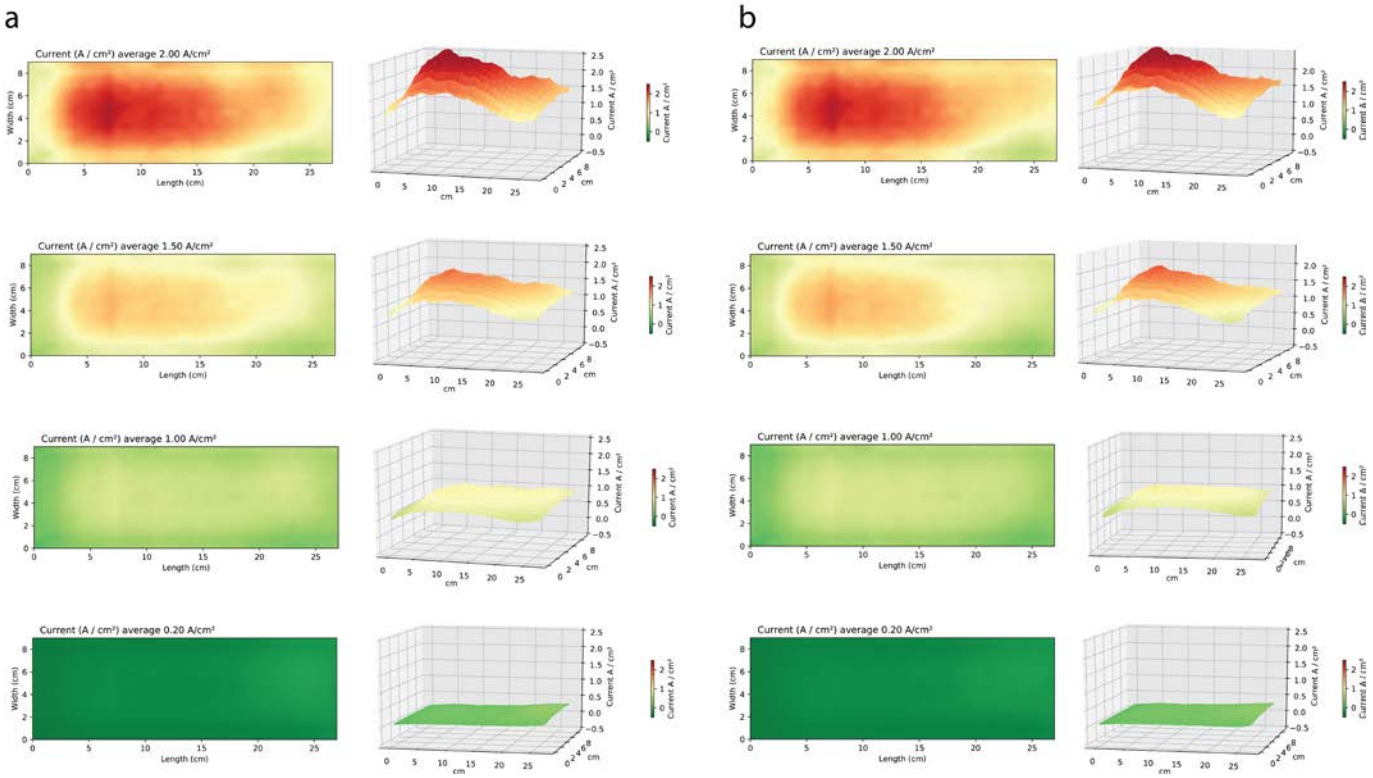

Figure 2 – Representative current distribution reads at 2D and 3D plot of tested regular MEA type #3 at different load points of 2.0, 1.5, 1.0, and 0.2 A cm^{-2} of the tested MEA type #3 that was in contact with the CDP. The left side of the plot is the air inlet and H_2 outlet region, and right side of the plot is the air outlet and H_2 inlet region. a) shows the current distribution at 70 °C, b) shows current distribution at 80 °C. The shows a gradient current behaviour at high loads result by mass transfer due to depletion of O_2 concentration gas.

Gradient MEA

The platform build consisted of regular MEAs type #1, MEAs type #2, and gradient MEA type #4 (which was facing the CDP), Table 5 presents the parameters of the MEA types that were in the second PEMFC build. The configuration of the gradient MEA has an average Pt load of 0.1 $mg_{Pt}\ cm^{-2}{}_{Pt}$, and a membrane thickness of 10 µm is similar to the regular MEA type #3. However, the loading of the gradient MEA consisted of a linear gradient from 0.05 $mg_{Pt}\ cm^{-2}$ at the air inlet, to and 0.15 $mg_{Pt}\ cm^{-2}$ at the outlet.

Table 5: Parameters of the MEAs that were in the second short-stack PEMFC build.

MEA type #	Average cathode loading $mg_{Pt}\ cm^{-2}{}_{Pt}$	Membrane thickness µm	Type CCM
MEA #1	0.4	15	Homogenous
MEA #2	0.1	15	Homogenous
MEA #4	0.1	10	*Gradient*

The test runs with gradient MEAs were similar to regular MEA test run, and consisted of polarisation curves recorded at 70 °C and 80 °C (Figure 3). Similar to the regular MEA, temperature had a minor effect on the performance. No effect of the temperature was observed in the kinetic region. At the ohmic region, the high loading MEA type #1 had negligible effect from the temperature change, whereas the low loading MEA type #2 had

a small but notable effect on the ohmic region and which slightly lowered the performance at 80 °C, compared to 70 °C.

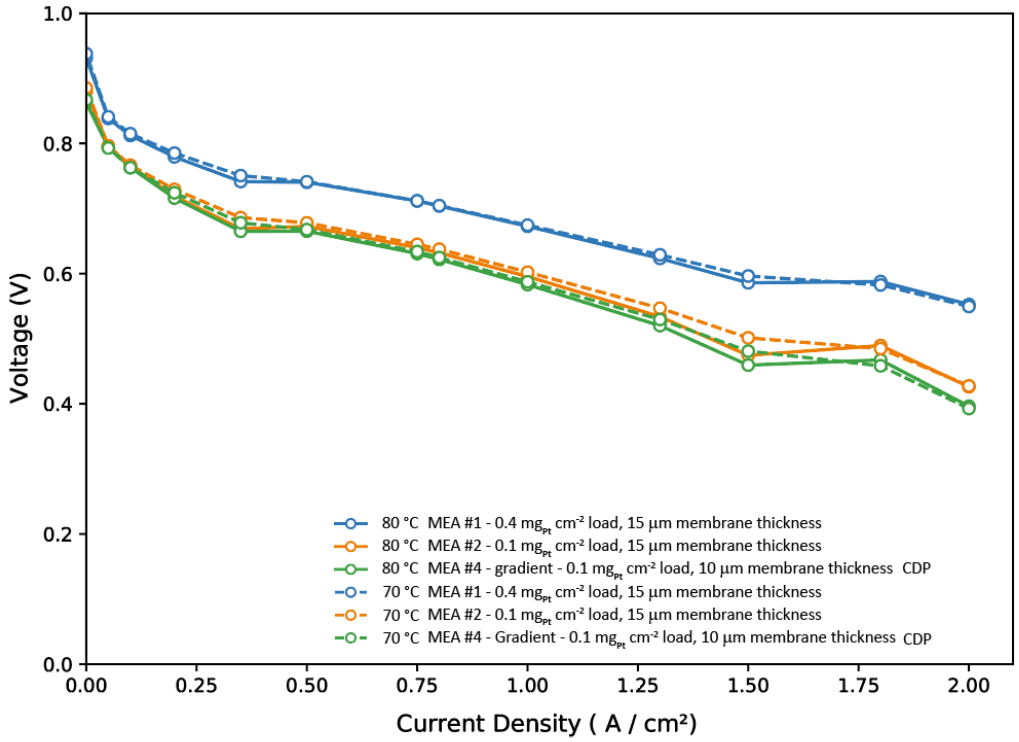

Figure 3 – The polarisation curves of three different types of MEAs that were tested simultaneously in the PEMFC stack. MEA #4, with average low loading of 0.1 mg$_{Pt}$ cm^{-2}, which was placed so that it was in contact with the current distribution plate (CDP).

Figure X shows snapshots of the 2D and 3D current distribution maps collected by CDP in contact with the gradient MEA type #4. Figures Xa and Xb show the typical current distribution activity of the gradient MEA #4 at 70 °C and 80 °C, at selected different load points (2.0, 1.5, 1.0 and 0.2 cm^{-2}). Similarly to the previous platform run, the first 1-2 cm of the active area was blocked by the gasket.

The first row shows the load of 2 A cm^{-2}, corresponding to the mass transfer region. At 70 °C, the current distribution is even across the active area, whereas at 80 °C, there is a slight gradient in current from inlet to outlet. The second row shows a similar homogenous current distribution at 1.5 A cm^{-2}. Compared to regular MEA with homogenous loading, the gradient MEA showed a gradient decline in performance. At lower current densities of 1.0 and 0.2 A cm^{-2} , there is higher activity close to air outlet due to higher catalytic loading. The behaviour of the gradient MEA is reversed compared to regular MEAs, where at lower current densities the current is homogenous, while at higher current densities, there is decrease in current from inlet to outlet.

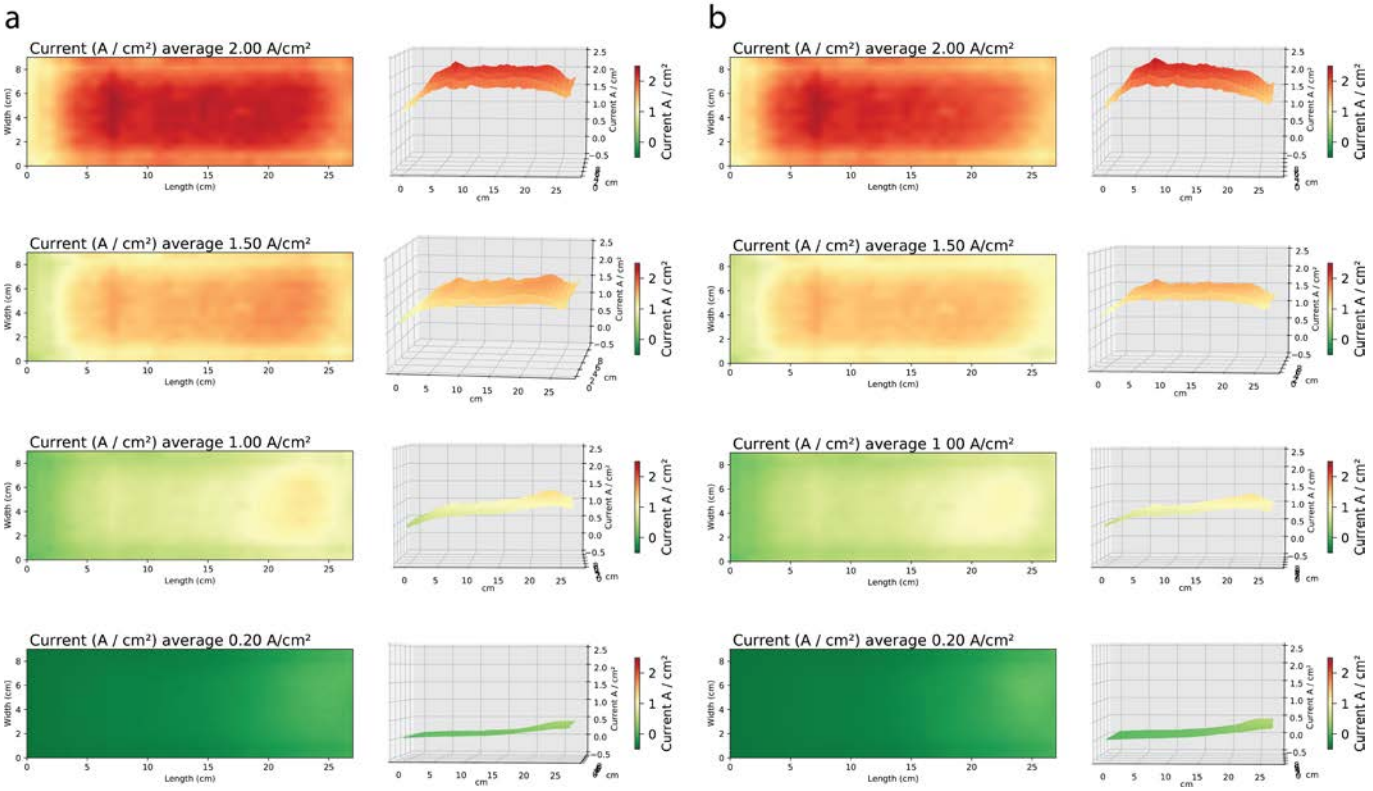

Figure 4 – Representative current distribution reads at 2D and 3D plot of tested gradient MEA type #4 at different load points of 2.0, 1.5, 1.0, and 0.2 A cm^{-2} of the tested MEA type #3 that was in contact with the CDP. The left side of the plot is the air inlet and H_2 outlet region, and right side of the plot is the air outlet and H_2 inlet region. a) shows the current distribution at 70 °C, b) shows current distribution at 80 °C. The shows a gradient current behaviour at high loads result by mass transfer due to depletion of O_2 concentration gas.

The behaviour of the gradient MEA (type #4), where the current distribution is even across the active area at high current densities, could be explained by the low/high load at air inlet/out, respectively. Unlike the regular MEA, most of the oxygen is consumed at the beginning of the MEA and is slowly depleting towards the outlet, making it less likely for the oxygen molecules to interact with the Pt particles. The increase of the Pt load closer to the outlet counteracts this by increasing the likelihood of O2 reacting. Thus the gradient MEA have an even current distribution, especially when the current near the outlet would otherwise be limited by mass transfer at high current loads. At kinetic and low ohmic regions of the gradient MEA, polarisation curve shows a gradient behaviour with higher currents at the air outlet region with high load. Since there are no mass transfer issues and the concentration of the O2 is relatively homogenous across the active area, it is more likely that the oxygen reacts near the outlet where the load of Pt is the highest.

Nevertheless, while a gradient MEA configuration such as MEA #4 provides a homogenous distribution of the current distribution with no decrease of current density across the channels, there are still some disadvantages. Figure 5 shows the comparison between the regular MEA, the voltage values from the gradient MEA is generally lower. However, the gradient MEA shows better activity in the mass transfer region, and the open circuit voltage (OCV) is slightly higher than the regular MEA.

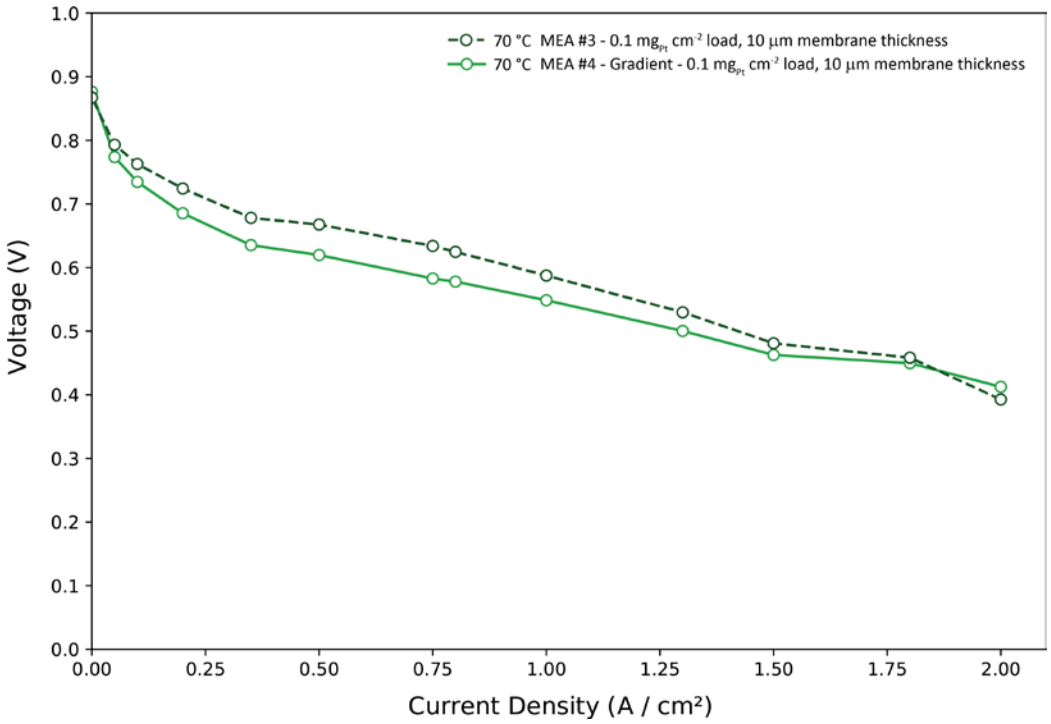

Figure 5 – The polarisation curves of two different MEAs regular MEA #3 and graidnet MEA #4 that were tested in contact with CDP. Both MEAs had similar membrane thickness of 10 µm, and average cathode load of 0.1 mg$_{Pt}$ cm^{-2}.

4 CONCLUSIONS AND FUTURE WORK

A new concept design of MEAs with gradient cathodic catalytic loading along the x-y axis was tested and compared to regular MEAs with homogenous cathodic catalytic load coating. Both types of MEAs have similar membrane thickness and similar average loading of the catalyst. The cathode loading for the regular MEA is 0.1 mg$_{Pt}$ cm^{-2}, whereas gradient MEA had a similar average loading of 0.1 mg$_{Pt}$ cm^{-2}, but the loading was varied linearly from 0.05 mg$_{Pt}$ cm^{-2} at the inlet to 0.15 mg$_{Pt}$ cm^{-2} at the outlet. Both MEAs were tested in a short-stack PEMFC equipped with CDP. Regular MEA showed expected behaviour with a gradient current distribution with decline performance across the channels, while gradient MEA showed an even current distribution. The gradient MEA of such configuration shows promising behaviour with improved mass transfer. Unfortunately, the trade-off comes in lower performance, which may be due to low loading, however, the benefit can come with better durability and reduced degradation rate compared to regular MEA due to gradient MEA being able to achieve to homogenous current distribution behaviour across the active area. Nevertheless, such MEA concepts are still in the prototype phase, and after optimisation, the activity could be better than regular MEA with similar average loading. Hence, it can reduce the cost per energy.

The future work will continue by using CDP and conducting experiments on MEA configurations with gradient catalytic loading in the Z axis alongside with x-y gradient, such MEA concepts may show promising results, with potential for future MEAs.

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6 APPENDIX

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