

# Correlation between Ex-situ and In-situ Contact Resistance of Bipolar Plates in PEMFCs

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# Outline

#### Introduction

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#### • Experimental

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#### • Results

Ex-situ contact resistance at room temperature Ex-situ contact resistance in simulated environment In-situ contact resistance



#### Introduction-contact resistance

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#### Contact resistance

- Porous nature of the GDL combined with the surface roughness of the BPP makes the effective contact area between these components crucial for the magnitude of the contact resistance
- When using uncoated steels as BPPs, the effective contact area is especially important due to the poorly conductive passive film on the surface of the steel

#### Maximization of the effective contact area

- ✤ Optimization of surface roughness of BPP<sup>1</sup> and GDL
- ✤ Young's modulus of GDL
- Compression pressure

(1) J. Andre', et al., International Journal of Hydrogen Energy 34 (2009) 3125–3133



### Introduction-Evaluation of contact resistance

#### Ex-situ measurements of contact resistance

- Relatively inexpensive and practical way to perform fast screening among candidate BPPs
- Commonly carried out at room temperature and in air atmosphere and may fail to predict possible changes in contact resistance due to the fuel cell environment

#### In-situ testing

- Essential to evaluate the initial performance, as well as the long term stability of the fuel cell
- When comparing different BPPs, the differences in fuel cell performance not always correlate to the differences in contact resistance <sup>2</sup>
- Changes in contact resistances can be very difficult to identify from other ohmic losses such as membrane resistance or electrode ionomer resistances<sup>3</sup>
- Post-mortem contact resistance measurement do not always correlate with observed changes in cell voltage<sup>4</sup>
- (2) Kai Feng, Tao Hu, Xun Cai, Zhuguo Li, Paul K. Chu, Journal of Power Sources 199 (2012) 207-213
- (3) M. Kumagai, S.-T. Myung, T. Ichikawa, H. Yashiro, Journal of Power Sources 195 (2010) 5501–5507
- (4) S. Auvinen, T. Tingelo, J. K. Ihonen, J. Siivinen, M. Johansson, Journal of The Electrochemical Society, 158 (5) B550-B556 (2011)







### Introduction-Evaluation of contact resistance

#### In-situ measurement of contact resistance

- Makkus et al. (2000)<sup>5</sup> measured the in-situ contact resistance of seven different stainless steels, among them 316L.
- Anode side stainless steel plates has lower contact resistances than the cathode side plates.
- Cathode side contact resistance increase during operation
- Ihonen et al. (2002)<sup>6</sup> fuel cell design that was able to separate the clamping pressure from the sealing pressure.
- A slight decrease in contact resistance with increasing current density, due to heat production at the cathode.
- An increase in contact resistance with increasing gas pressure, where new surface is exposed to oxide growth.
- Contact resistance may grow to unacceptable levels when cycling the clamping pressure or the gas pressure.





(5) R. C. Makkus, A. H.H. Janssen, F. A. De Bruijn, R. K. A. M. Mallant, J. Power Source 86 (2000) 274-282
(6) J. Ihonen, F. Jaounen, G. Lindbergh, G. Sundholm, Electrochimica Acta 46



## Experimental-setup

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#### Ex-situ simulated environment

In-situ



*Bi-polar plate samples (area:7 cm<sup>2</sup>)* 

- Modified version of Ihonen's fuel cell design
- Active area and flow field of 7 cm<sup>2</sup> (spiral flowfield)
- Land area: 3.86 cm<sup>2</sup> (graphite) and 3.82 cm<sup>2</sup> (stainless steels)
- Channel depth: 1 mm (graphite) and 0.4 mm (stainless steels)
- Each BPP sample is tested in all the three setups



## Results of ex-situ measurements

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#### Ex-situ

*Evaluation of the contact resistance* at room temperature and the effect of GDL

Conducti



# Ex-situ contact resistance at room temperature

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*Conditions: GDL: Sigracet 10BC Current density: 0.14 A cm<sup>-2</sup> Temperature:23 °C (room temperature)* 

- The conducting properties of the materials, as well as the effective contact area are crucial for the magnitude of contact resistance
- Contact resistance of uncoated 316L is approximately one order of magnitude larger than graphite
- Increasing clamping pressure compresses and deforms the GDL, increasing the effective contact area between BPP and GDL
- Every contact spot is much more important for poorly conducting BPP materials



#### Effect of GDL



a)

b)

1.0kV 8.4mm x130 SE(M



# Results of ex-situ measurements in simulated environment



#### Ex-situ in simulated environment

Isolate the effect of current density, temperature and humidity without the influence of the MEA, water and heat production at the cathode



## Effect of current density

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*Conditions: GDL: Sigracet 10BC Temperature:23 °C (room temperature)* 

- After each current step the voltage drop between GDL and BPP (contact resistance) decreases with time and it is more pronounced at low clamping pressures.
- The cell temperature also increases with increasing current density. This is due to the thermal resistances and it is also more pronounced at low clamping pressures.
- The contact resistance dependence on current density might be an effect of temperature.



# Effect of temperature

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*Conditions: GDL: Sigracet 10BC Current density: 0.14 A cm<sup>-2</sup> Clamping pressure: 8 bar (80 Ncm<sup>-2</sup>)* 

- All three materials show a strong dependence on temperature, where the contact resistance decreases with increasing temperatures
- ✤ Changes in electronic conductivity
- Thermal expansion of the materials is probably the main contributor to the contact resistance decrease.

Increase the internal pressure of the cell, deforming the GDL and enlarging the effective contact area between GDL and BPP

• The contact resistance is not completely reversible, just as the structure of the GDL upon compression



# Effect of humidity/water

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- The introduction of oversaturated gases increases the pressure drop over the flow field/BPP (lower for graphite due to deeper channels)
  - Large changes in pressure drop seems to change the effective contact area between GDL and BPP, increasing the contact resistance.
- For uncoated 316L both dynamic and permanent changes in contact resistance are observed



### Results of in-situ measurements



 <u>In-situ</u>
 *Effect of temperature, humidity, current density* and time



# Effect of temperature

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- Considerable differences in cell voltages. HFR-corrected cell voltage show that ohmic losses are indeed the largest contribution to this difference
  - The HFR is one order of magnitude larger than the contact resistance
- While the HFR show an exponential increase, the contact resistance shows a clear decrease. Probably due to thermal expansion of the materials 7
- The large differences in cell voltage are not due to the contact resistance, as the difference in HFR is much larger than the difference in contact resistance

#### Conditions:

MEA: Gore 5621 GDL: Sigracet 10BC Current density: 0.14 A cm<sup>-2</sup> Clamping pressure: 8 bar (80 Ncm<sup>-2</sup>) Temperature: 80 °C Gas: Dry O<sub>2</sub> and H<sub>2</sub> Flow rate: 60 ml/min for both O<sub>2</sub> and H<sub>2</sub>  The cell operated with uncoated 316L has much shallower flow field channels, causing a higher gas velocity and having a drying effect on the membrane

(7) Y. Zhou, G. Lin, A.J. Shih, S.J. Hu, Journal of Power Sources 192 (2009) 544–551



# Effect of humidity

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- Similar cell voltages between uncoated 316L and graphite despite existing differences in contact resistance. Uncoated 316L may have slightly better electrode performance due to differences in over night conditioning
  - A near exponential decrease of the HFR as the ionic conductivity of the membrane and electrodes is improved.
  - A fraction of the HFR decrease is due to the contact resistance.
- ✤ A possible contribution from membrane swelling with increasing RH<sup>7</sup>
- However, the contact resistance of uncoated 316L steel stops to decrease at 50 % RH and instead increases at high water contents and high pressure drops at the cathode

#### Conditions:

MEA:Gore 5621

- GDL: Sigracet 10BC
- Current density: 0.14 A cm<sup>-2</sup>
- Clamping pressure: 8 bar (80 Ncm<sup>-2</sup>)
- *Temperature:* 80 °C
- Gas: humidified  $O_2$  and  $H_2$
- Flow rate: 60 ml/min for both  $O_2$  and  $H_2$

(7) Y. Zhou, G. Lin, A.J. Shih, S.J. Hu, Journal of Power Sources 192 (2009) 544–551



# Effect of current density

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- An increase in voltage drop between the BPP and GDL at high current densities, for all three samples
- Increase in temperature and pressure drop
- At 0.71 A cm<sup>-2</sup> contact resistance increases 0.41 m $\Omega$  cm<sup>2</sup>, 1.4 m $\Omega$  cm<sup>2</sup> and 11 m $\Omega$  cm<sup>2</sup> for graphite,, gold-coated and uncoated steel, respectively.





# In-situ contact resistance during long term operation



#### Conditions:

MEA: Gore 5621 GDL: Sigracet 10BC Current density: 0.14 A cm<sup>-2</sup> Clamping pressure: 8 bar (80 Ncm<sup>-2</sup>) Temperature: 80 °C Gas:  $O_2$  and  $H_2$  (100 %RH) Flow rate: 60 ml/min for both  $O_2$  and  $H_2$ 

- After each performed polarization curve the contact resistance changes to a new level, especially for the cell using uncoated 316L steel
  - Despite the changes in contact resistance the cell voltage improves during the 100 h period for all three samples
  - The contact resistance of uncoated steel 316L increases to 300 m $\Omega$  cm<sup>2</sup> after extensive time at high current density operation (+800 h)



## Conclusions

- The effective contact area between GDL and BPP is crucial for the magnitude and stability of the contact resistance, especially for poorly conductive materials such as uncoated stainless steels
- The contact resistance decrease with increasing temperatures, probably due to thermal expansion of the fuel cell components.
- The contact resistance decreases with increasing RH of the gases, probably due to membrane swelling. On the other hand, a large increase in pressure drop due to the presence of liquid water affects the contact resistance negatively.
- Current density has a dual effect on the contact resistance. While an increased current density increases the temperature of the cell and therefore have a positive effect on the contact resistance, it may also increases the water production and the pressure drop at the cathode, having a negative effect on the contact resistance.
- The changes in contact resistance could easily have been mistaken for other ohmic losses, such as metal ion poisoning of membrane, if the direct measurement of the contact resistance would have not been carried out.



• Thank you for the attention!