Investigation of Proton Exchange Membrane Fuel Cell Stacks for High Temperature Operation

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AUTOBRANE

(Automotive High Temperature Fuel Cell Membrane)

- Polymer electrolyte fuel cell stacks assembled with Johnson Matthey Fuel Cell and SolviCore MEAs based on the Aquivion™ (Solvay Solexis) short-side chain, chemically stabilized perfluorosulfonic acid membrane were investigated in the framework of the Autobrane EU FP6 Project;

- Electrochemical experiments in fuel cell short stacks were performed under automotive applications at pressures of 1-1.5 bar abs. over a wide temperature range, up to 130 °C, with varying levels of humidity (down to 18% R.H.);

- The short stacks were based on a Nuvera hardware properly implemented for high temperature operation and consisted of 6 large area (360 cm^2) MEAs;

- The membrane consisted of a thin (30μm) short side chain perfluorosulfonic polymer with high ion exchange capacity (equivalent weight of 790 g/eq) and suitable crystalline characteristic allowing proper mechanical properties in the overall temperature range.

Objectives
(Autobrane DOW)

Demonstration in a state of the art stack adapted to higher temperature demands of the membrane-electrode-assembly technology developed within the Autobrane project

Demonstration of a proof of concept of the new membrane and MEA technology in a stack with realistic area and a power of 1 kW

Overview

- Objectives
- SP5000 activities
- Stack assembling
- Stack testing
- Conclusions, Comments and Questions

Objectives (Autobrane DOW)

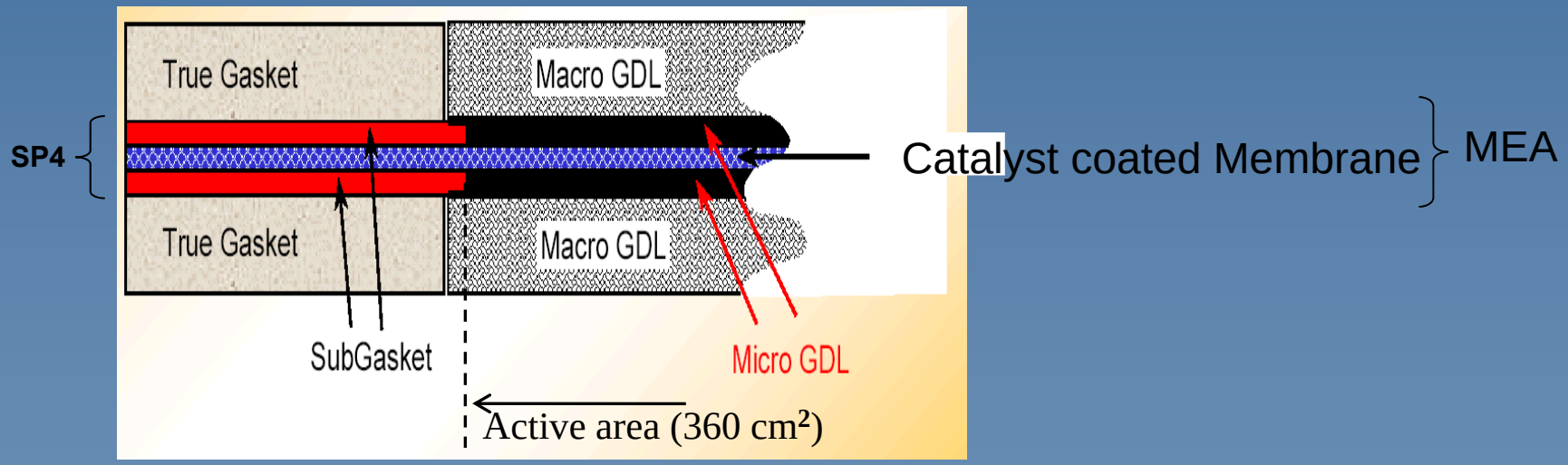
Operating specifications and conditions of the demonstrator

- Nominal power > 1kW
- Number of cells ~ 5
- Performance ~ 1 W cm^{-2}
- Cell size > 200 cm^2
- Stoich. H_2 ≤ 1.5, Air ≤ 2
- Temperature: up to 130 °C
- Pressure ≤ 1.5 bar abs.
- Low or internal humidification
- Operating mode including automotive relevant (dynamic)

Assembly and testing of short stacks

Dimensions of the cell active area and geometrical features

- Active area: 360 cm^2 (240mm x 150mm); this is the catalysed membrane area
- Total membrane area: 263.5 mm x 197 mm
- The membrane area that is not covered by the catalyst, is covered by the sub-gasket



MEA Thickness: 600-800 microns

➢T5140 - Assembly and testing of stacks

Preliminary conditioning of stacks and leak tests

Short stacks (JM and SolviCore): 6 cells

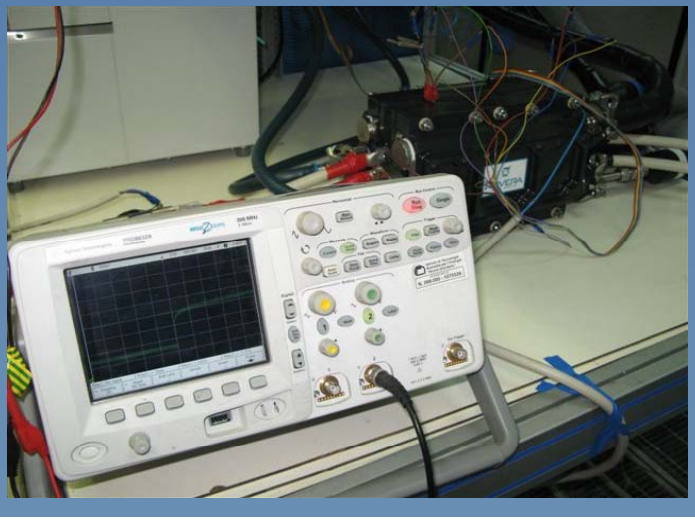
Stack compression: 10-18 kg / cm² (active area); electrode area: 360 cm²

SolviCore

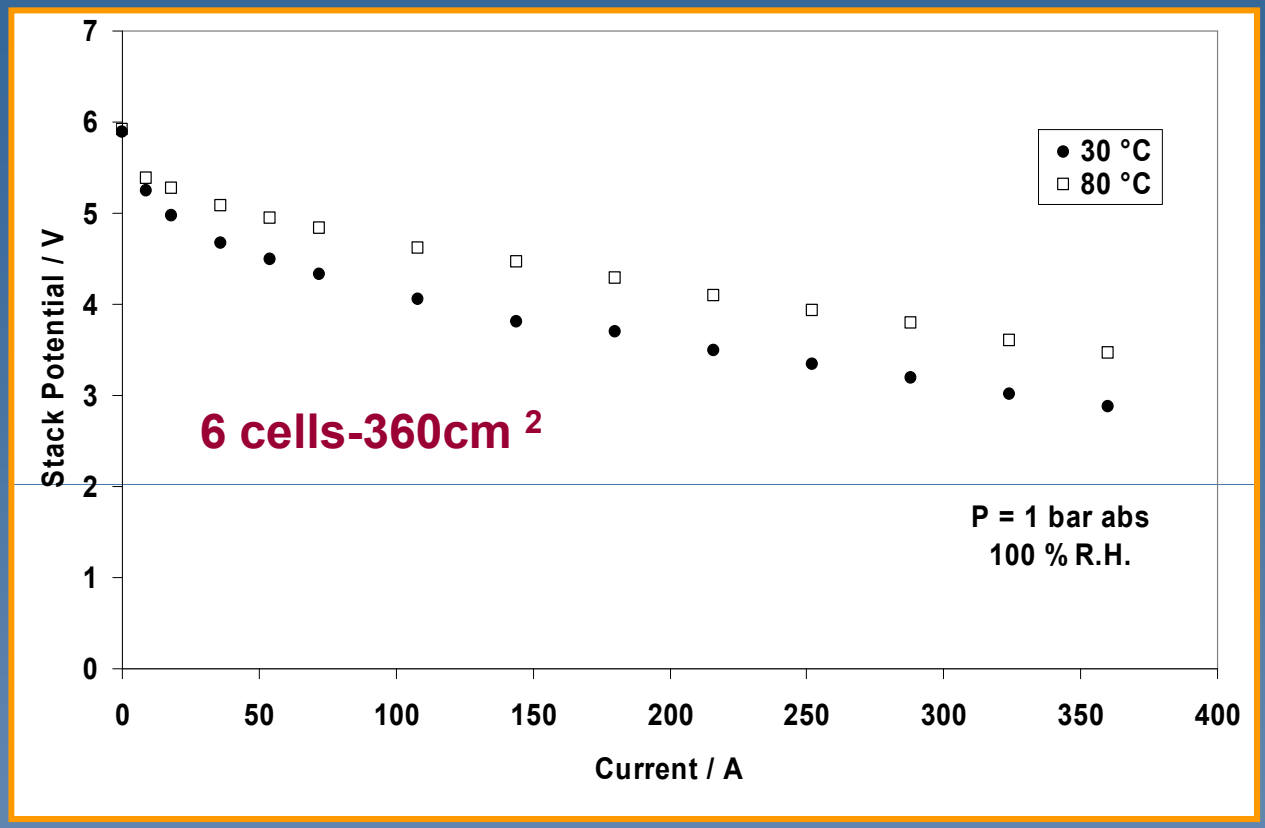
JM

Nuvera
Stack Hardware

Stack test station



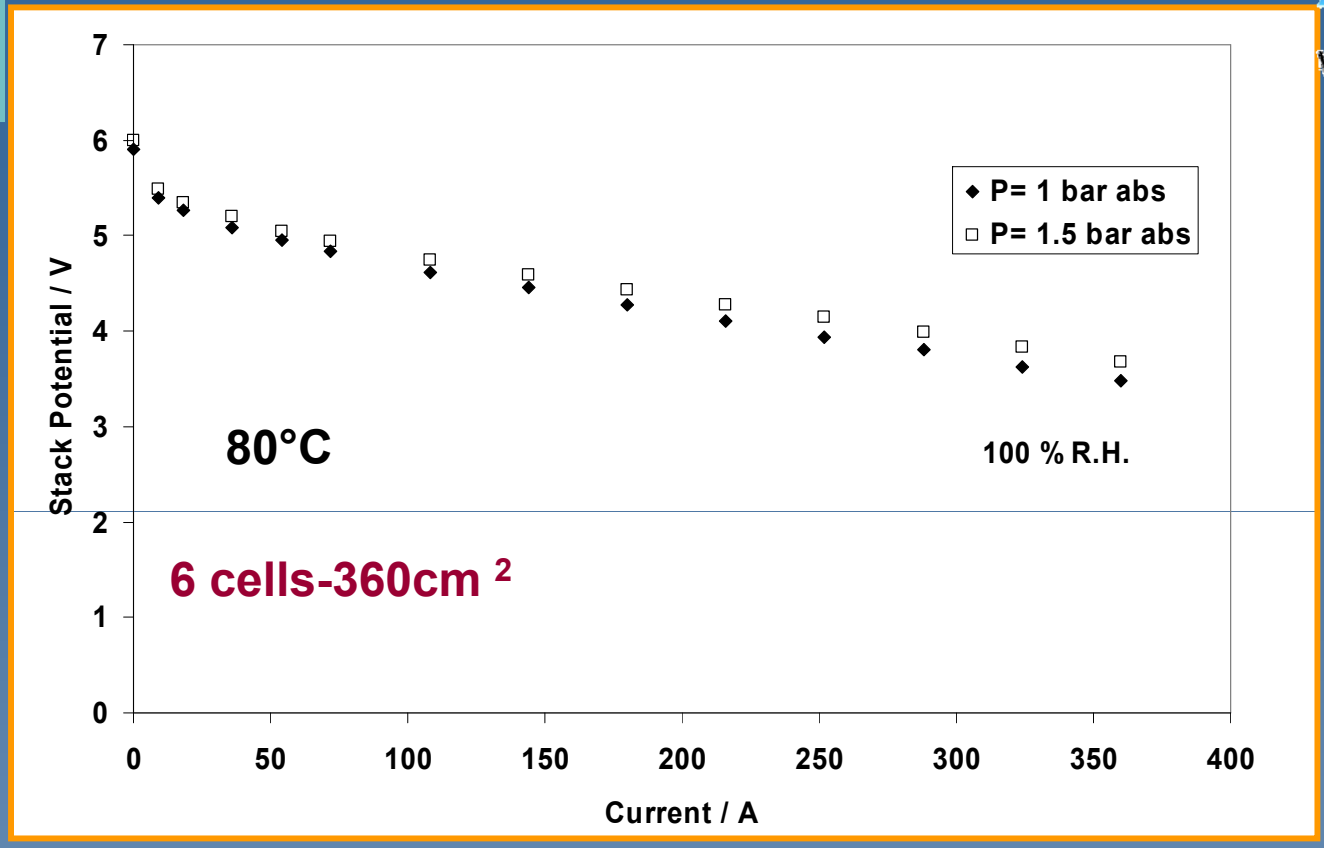
Low temperature performance



Polarization curves at two different temperatures for a 6-cells short stack assembled with JMFC 2nd generation MEAs (active area 360 cm2).

Comment: The stack showed appropriate performance at both low temperature and 80 °C. The combination of highly active electrocatalysts with a thin (30 µm) short side chain perfluorosulfonic membrane with super-acid properties (equivalent weight equal to 790 g/eq.) allows to enhance the reaction rate at low temperatures. These properties are useful for a rapid start-up of the device.

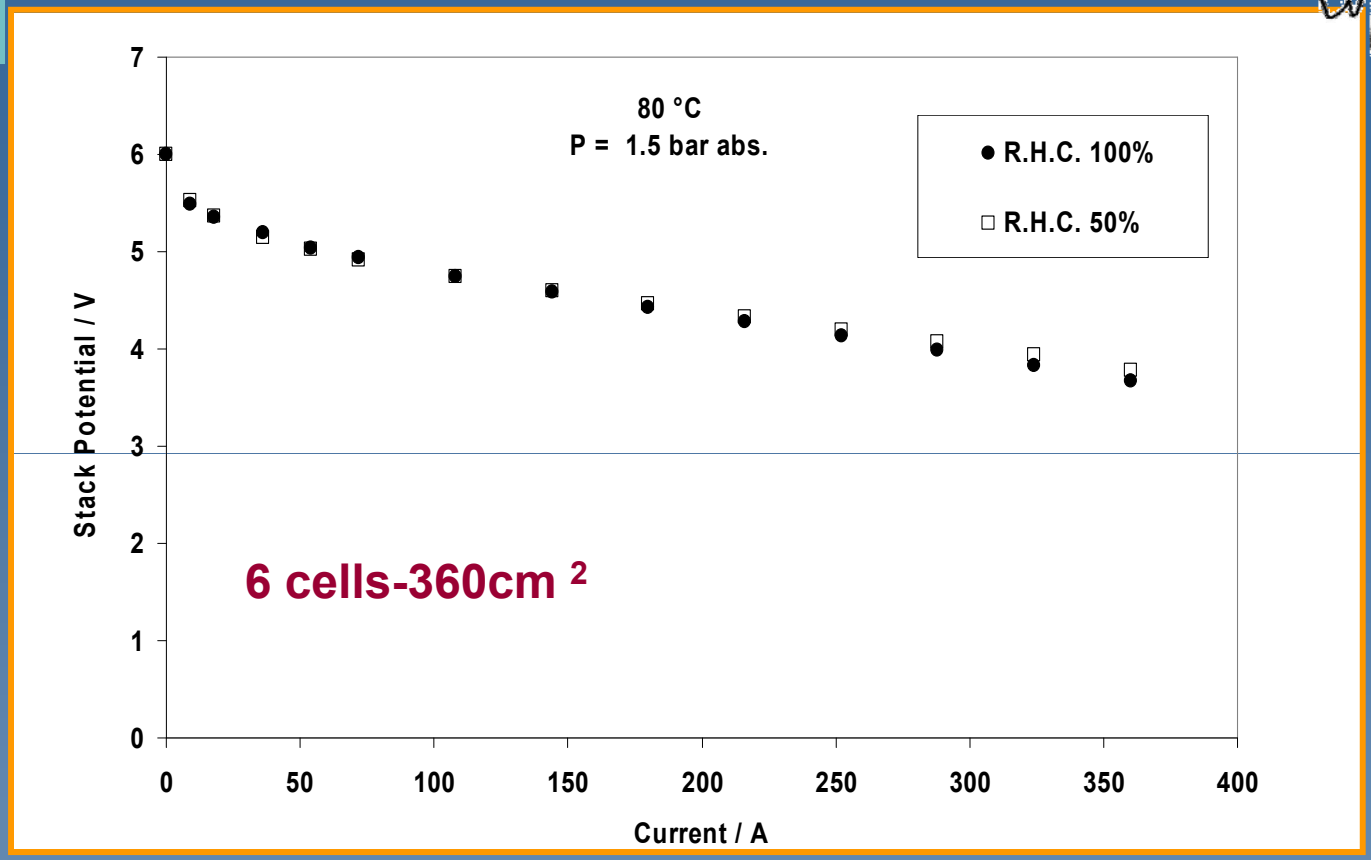
Effect of Pressure



Polarization curves at 80 °C and different pressures for a 6-cells short stack assembled with JMFC 2nd generation MEAs (active area 360 cm2).

Comment: A slight increase of performance is registered at 80 °C when the pressure is increased from 1 to 1.5 bar abs. An increase of pressure is especially useful at temperatures above 100 °C to maintain a small amount of liquid water inside the membrane; however, it can not exceed a certain limit which is determined by the technology of the air blowers for automotive applications presently available on the market. An air compressor is generally excluded from the automotive applications because of the high electrical power consumption and the large space needed to allocate this device on board with respect to the blower.

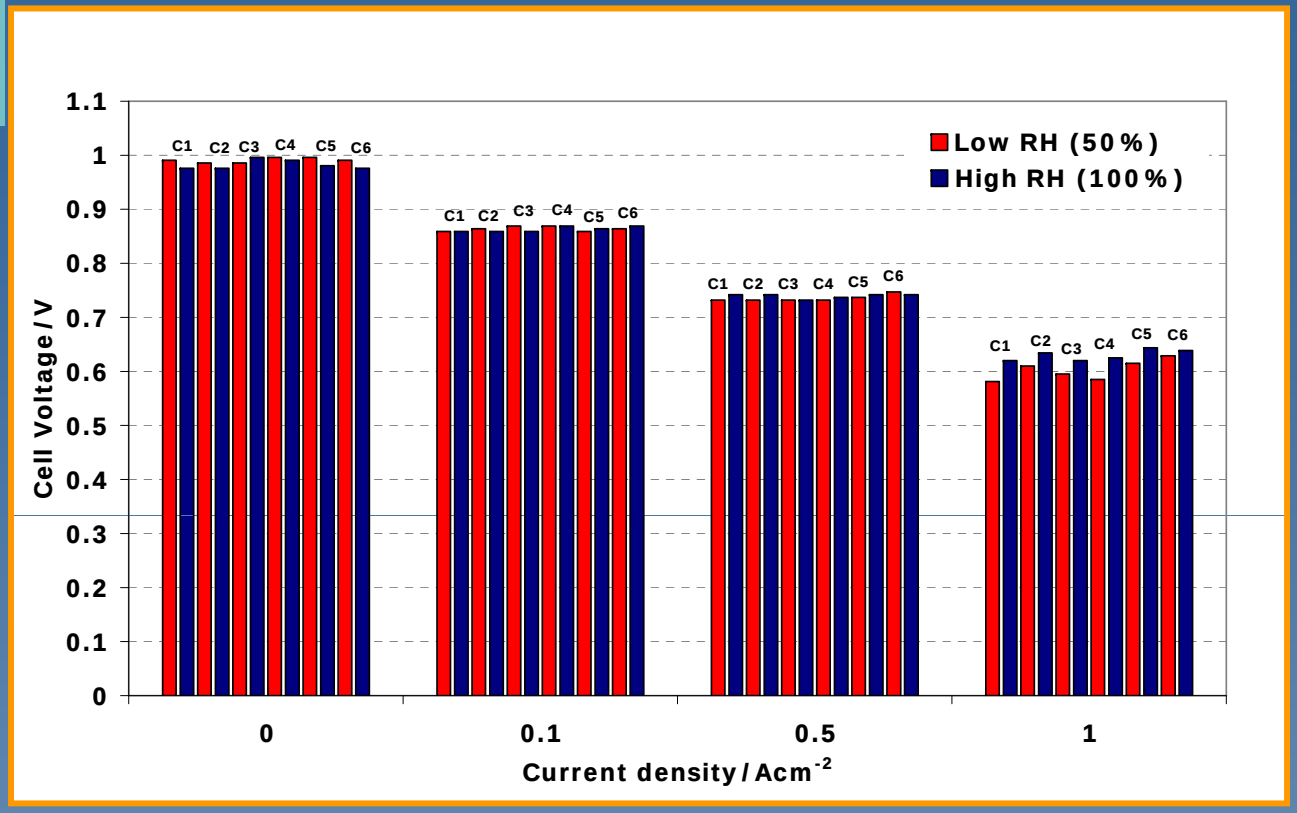
Effect of cathode relative humidity



Polarization curves at different cathode relative humidity for a 6-cells short stack assembled with JMFC 2nd generation MEAs (active area 360 cm2).

Comment: The slight increase of peak power density appears to be related to a lower flooding effect of the cathode under 50% R.H. at high current densities.

Distribution of Cell Voltages



Distribution of cell voltage at two levels of gas humidification:

JM short stack

R.H. A. =100% R.H. C.= 100%	R.H. A. =100% R.H. C.= 50%

Comment: All cells show the same behaviour with a small difference, at high current density. This behaviour was found to be related to a poor compression a larger contact resistances for the inner cells

- **T5240** - Performance tests
- **T5250** - Electrochemical tests

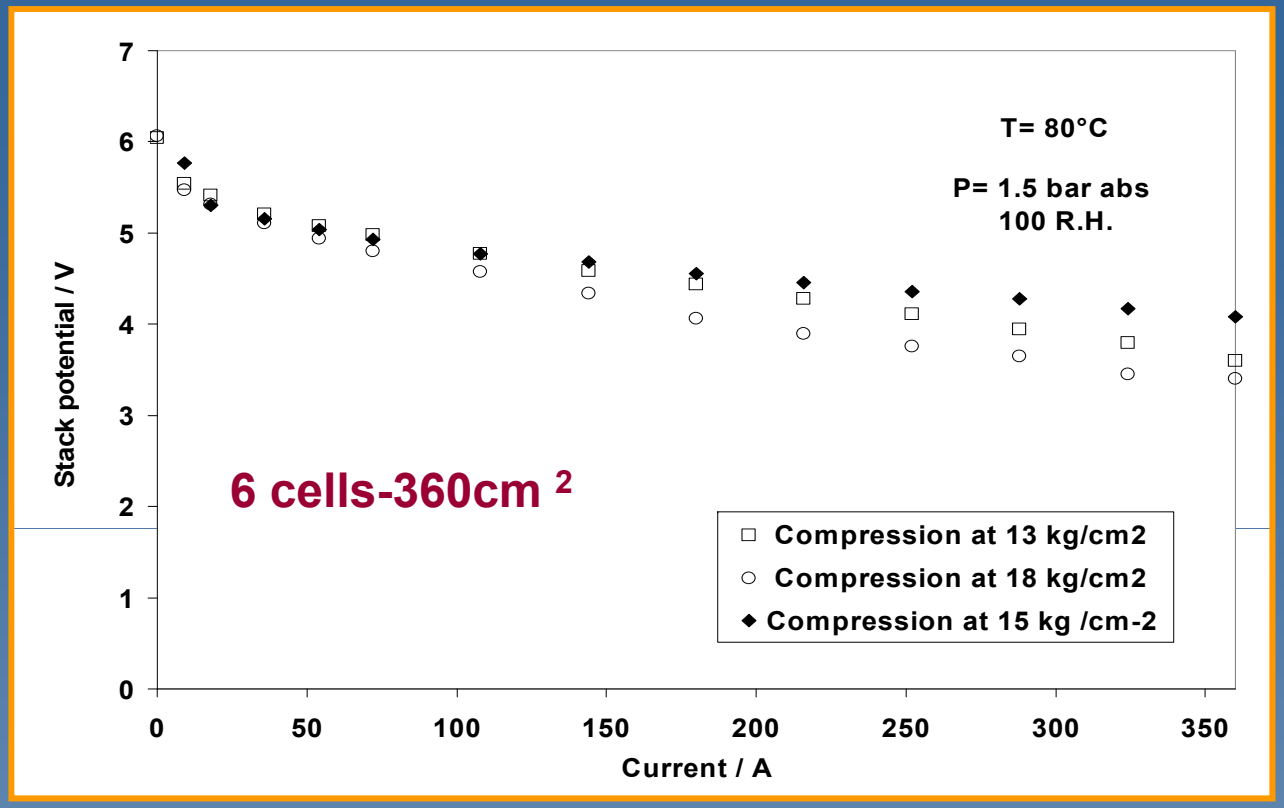
I=292.5 A V=3.9 V

I=364.6 A V=3.574 V

Power output larger than 1kW with 6 cells 360 cm2

Stack Temperature = 80 ° C
Pressure = 1.5 bar abs.
Compression = 13 kg cm^{-2}

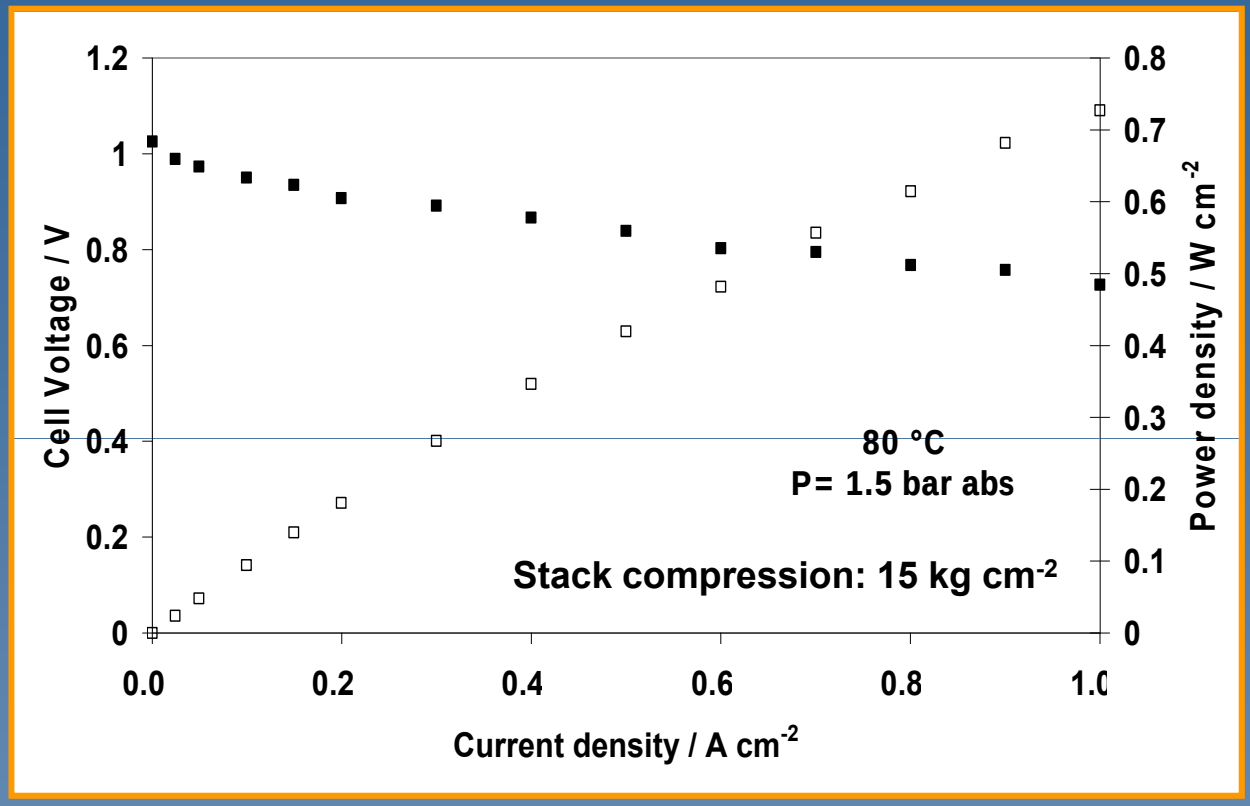
Effect of stack compression



Polarization curves for 6-cells short stacks assembled under different compressions with SolviCore MEAs (active area 360 cm^2).

Comment: A significant effect was played by the value of stack compression on the stack performance at low temperature. A high compression allows to achieve lower contact resistance but above a certain limit, a large pressure drop was observed which caused an increase of mass transport constraints. It is also pointed out that an over-compression also caused the occurrence of MEAs damaging. The optimum value of compression varied indeed for the different sets of MEAs; it is thus related to the MEAs characteristics as well as to the stack macroporous GDL/current collectors.

Best Normalised Performance under Conventional Conditions



Polarization and power density curves at 80 °C of a single cell in a stack assembled under optimised compression with Solvicore MEAs (active area 360 cm2)

Comment: The best performance was achieved at 80 °C with a peak power density of about 730 mW cm^{-2} (R.H.A. 100%, R.H.C. 50%, Air stoich. 2, H2 stoich. 1.5) in the presence of an optimised stack compression. *However, average recorded power densities under conventional conditions were in the range of 600 -700 mW cm^{-2} at an average cell voltage of 0.6-0.65 V.*

Conventional Operating Conditions Comments:

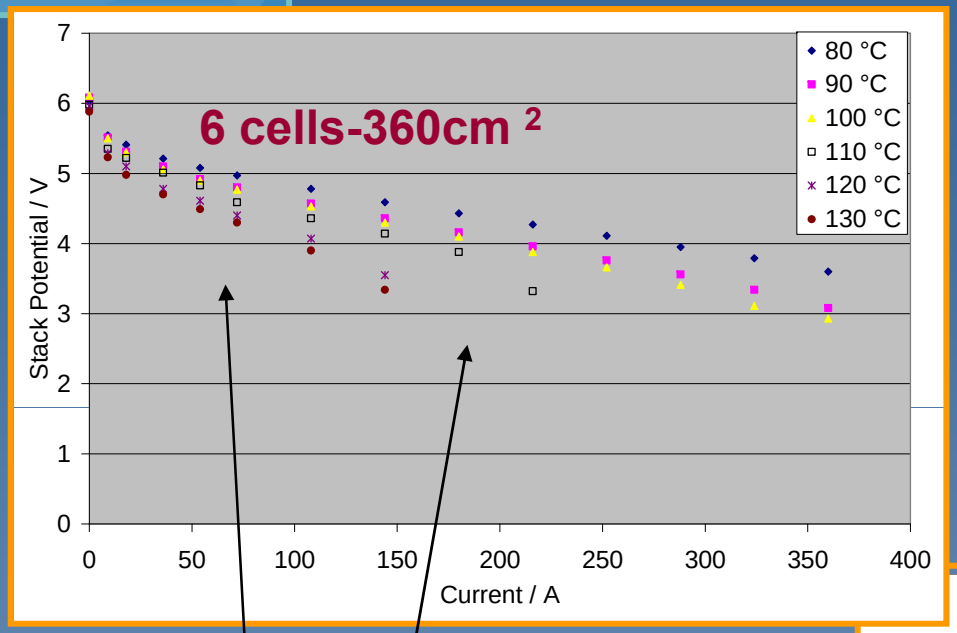
- The electrochemical behaviour of JMFC and SolviCore MEAs-based stacks was similar under conventional operating conditions.

- The average cell performance was between 600 and 700 mW cm^{-2} at an average cell voltage of 0.6-0.65 V at 80 °C.

- An output stack power better than 1 kW was achieved under conventional conditions with 6 cells-stack

- A significant effect is played by the value of stack compression on the performance especially at low temperature. An increase of compression may reduce ohmic limitations and thus increase the performance; however an increase of stack compression may result in MEAs damage. The optimization of compression is related to the MEAs characteristics as well as to the stack macroporous GDL/current collectors.

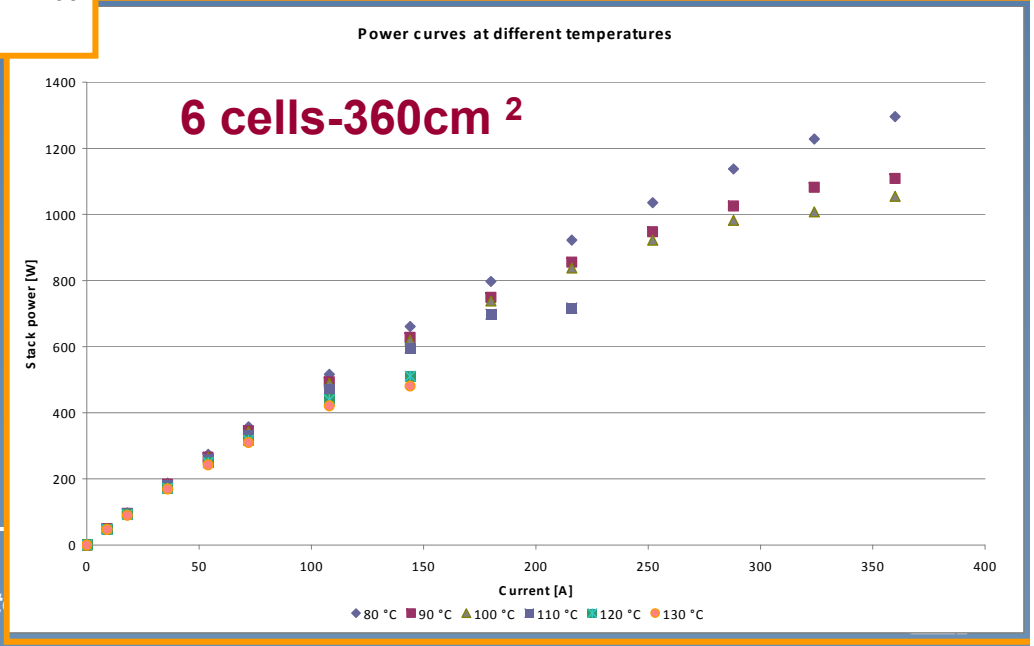
Effect of Temperature



SolviCore-MEAs based STACK

Compression 13 kg cm^{-2}

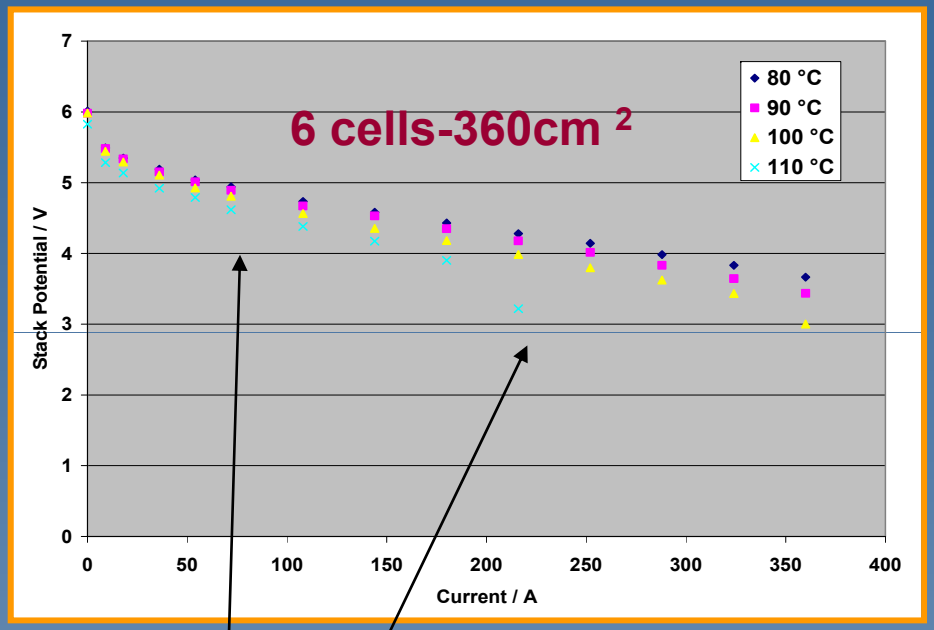
T umidifiers 80 °C
Pressure 1.5 bar abs.

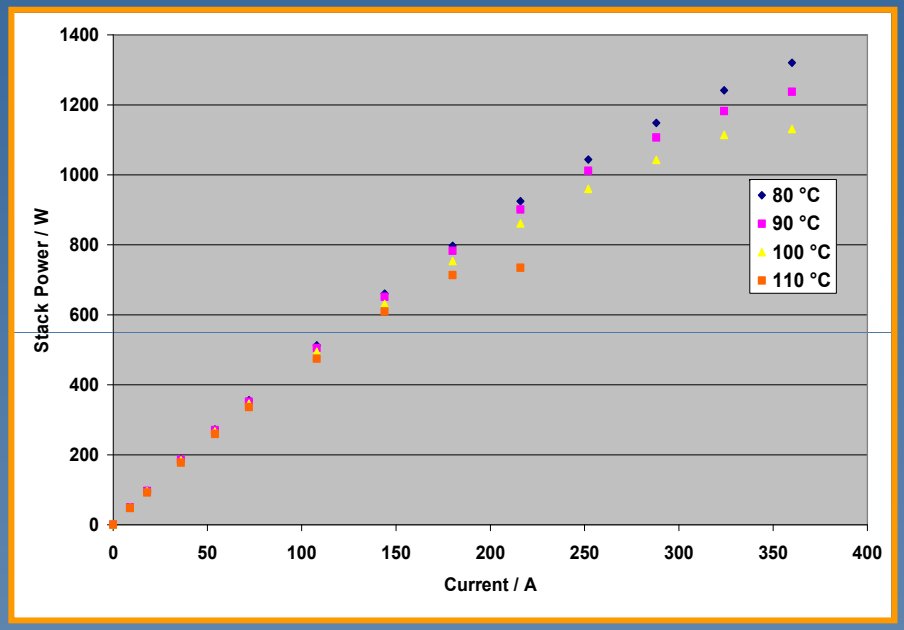


Evidence of Proton Transport Constraints at High Temperature

Effect of Temperature

JMFC-MEAs based STACK





Compression 13 kg cm^{-2}

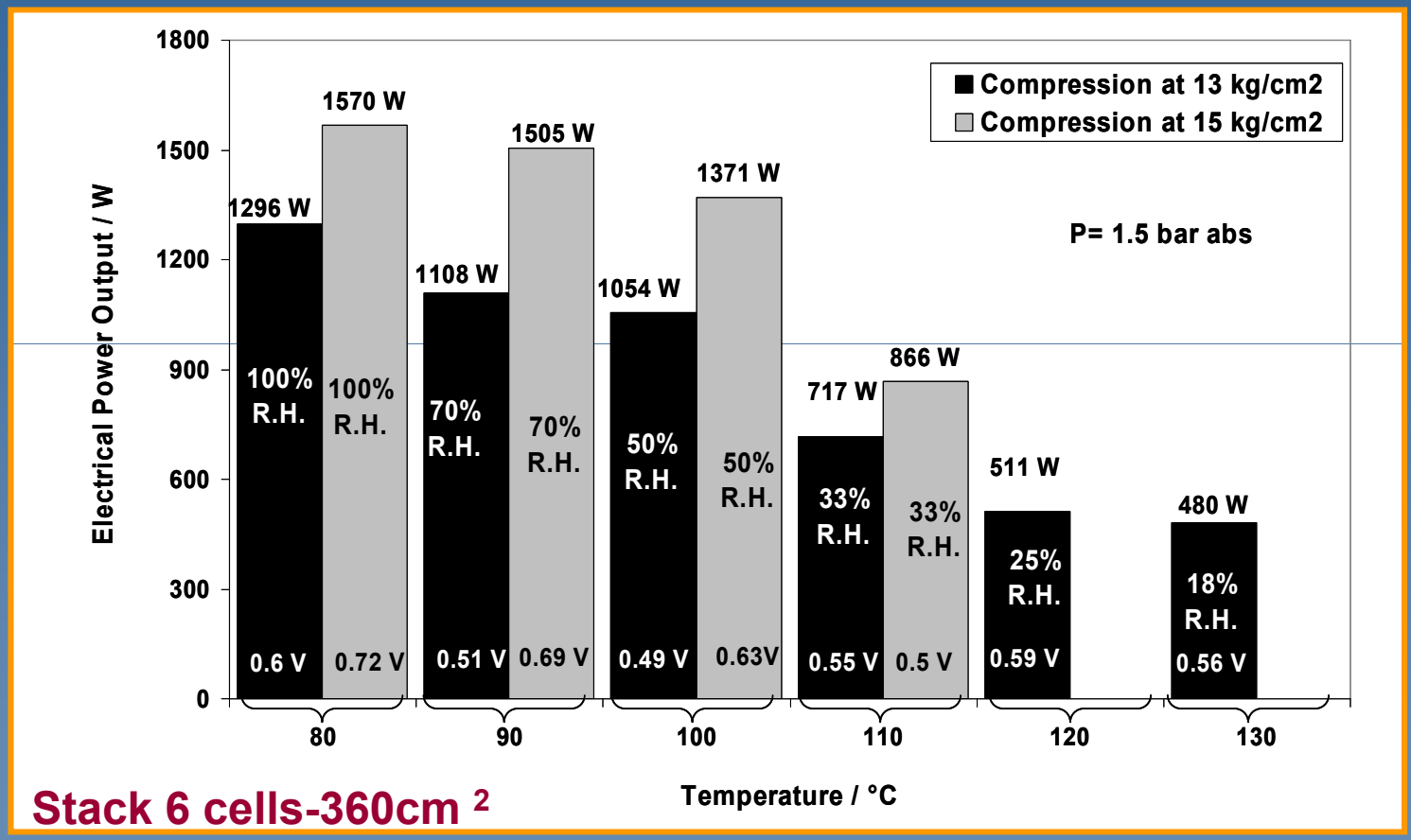
T umidifiers 80 °C
Pressure 1.5 bar abs.

Evidence of Proton Transport

Constraints at High Temperature

Solvicore peak power at two different compressions

2nd generation

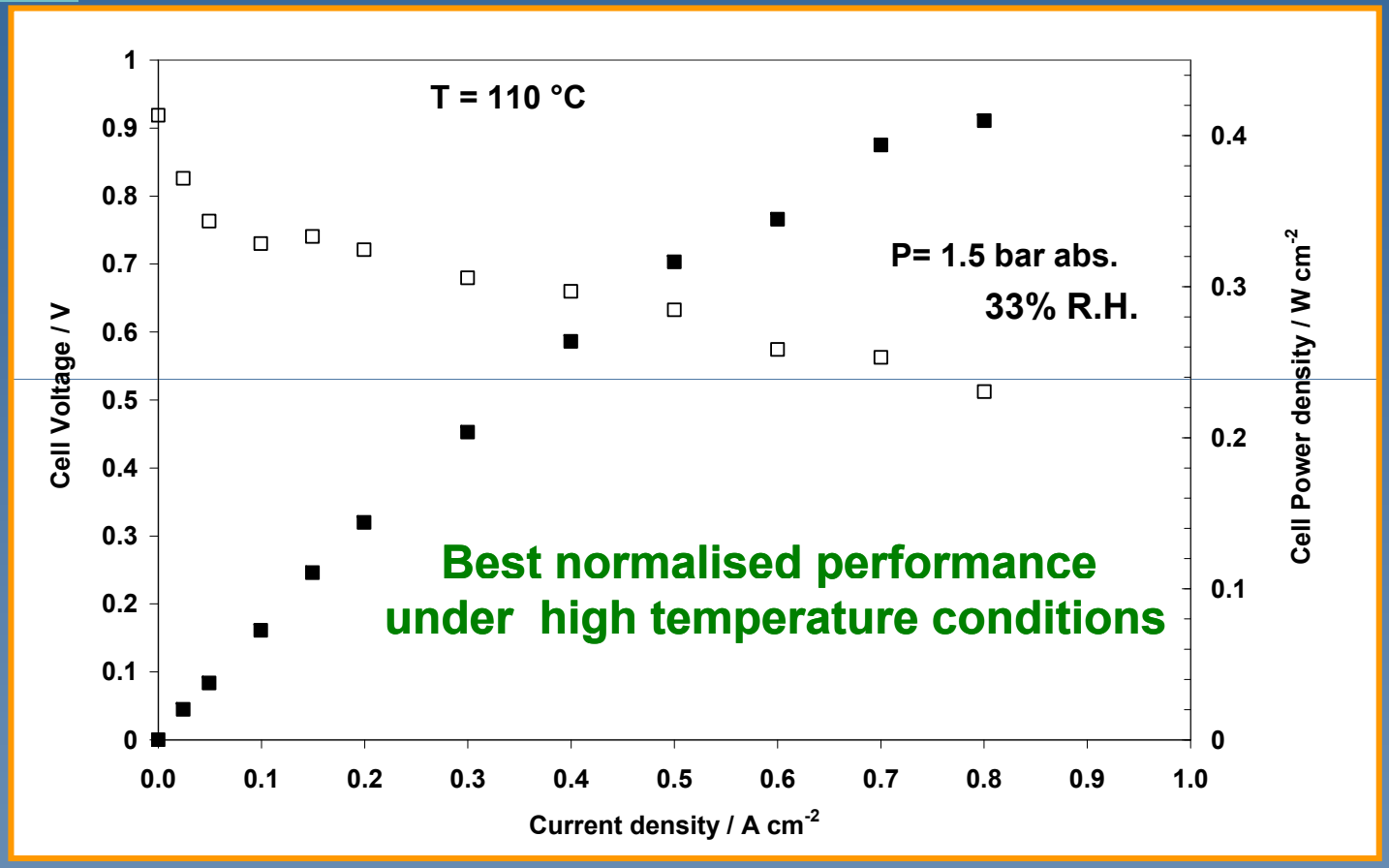


Stack 6 cells-360cm 2

Variation of the electrical power output as function of temperature for 6-cells short stacks assembled under two different compressions with Solvicore MEAs (active area 360 cm2).

SolviCore MEAs-based stack

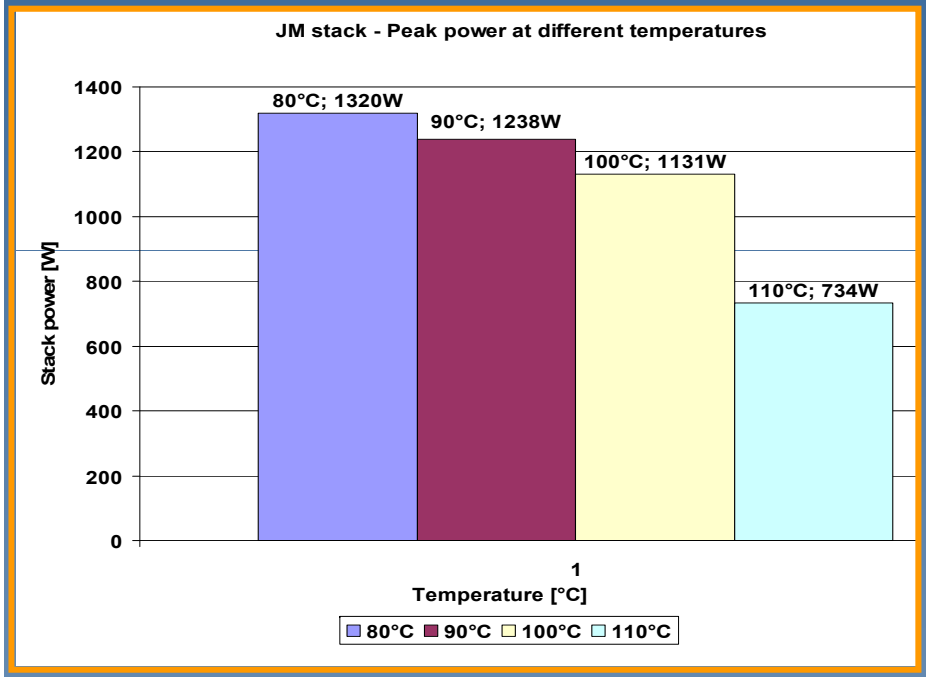
2nd generation

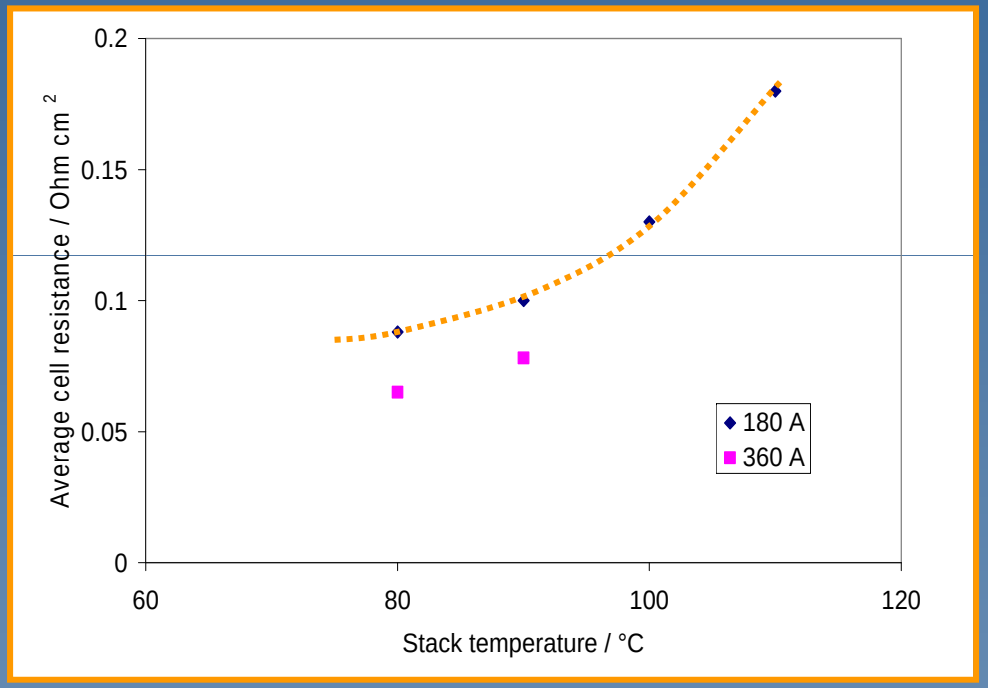


Operating Temperature/ Pressure
110 °C/ 1.5 bar abs. / 33 % R.H.

JMFC Peak Power and average cell resistance at different temperatures

2nd generation



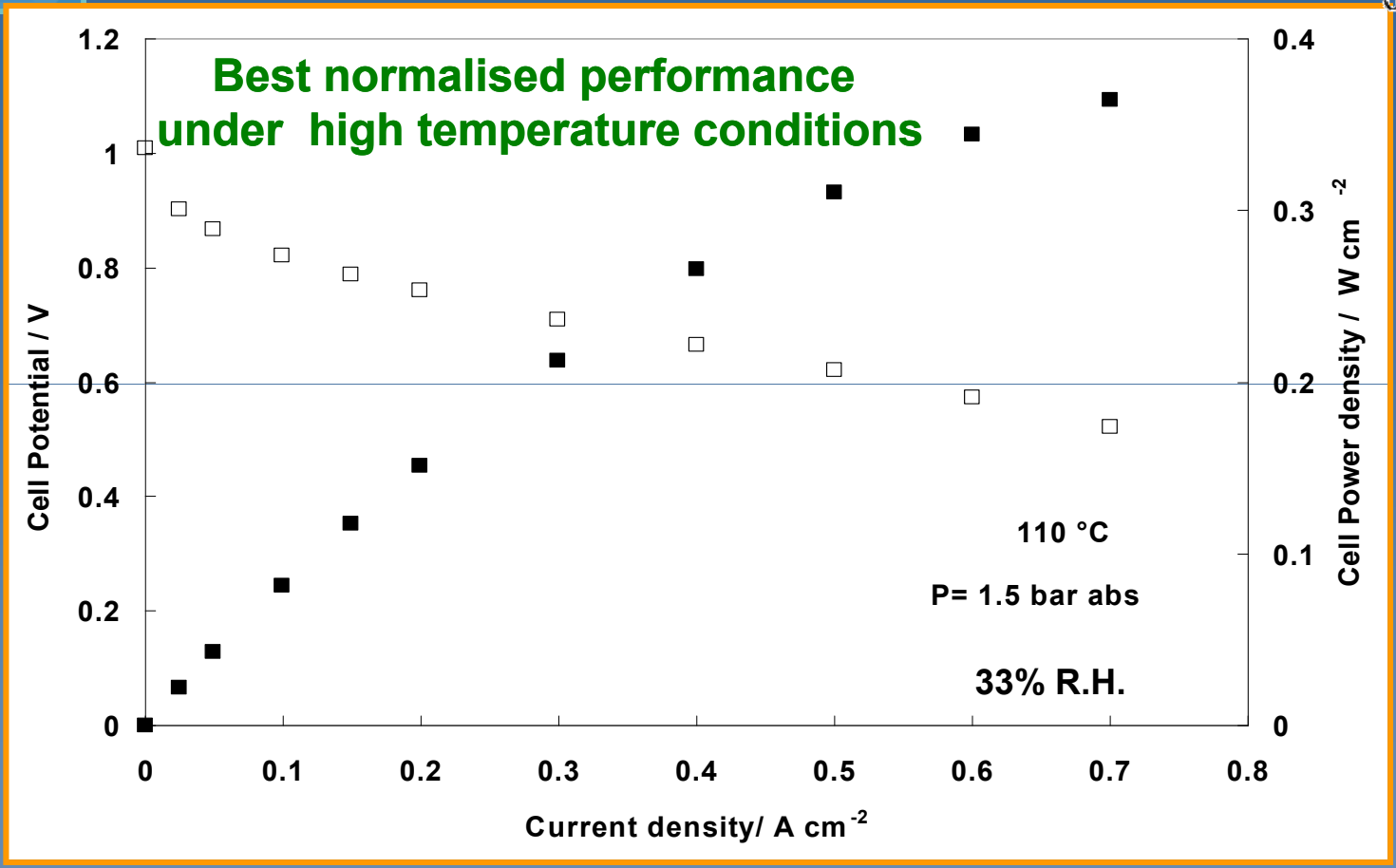


13 kg cm^{-2}

Stack 6 cells-360cm ^{2}

JM MEAs-based stack

2nd generation

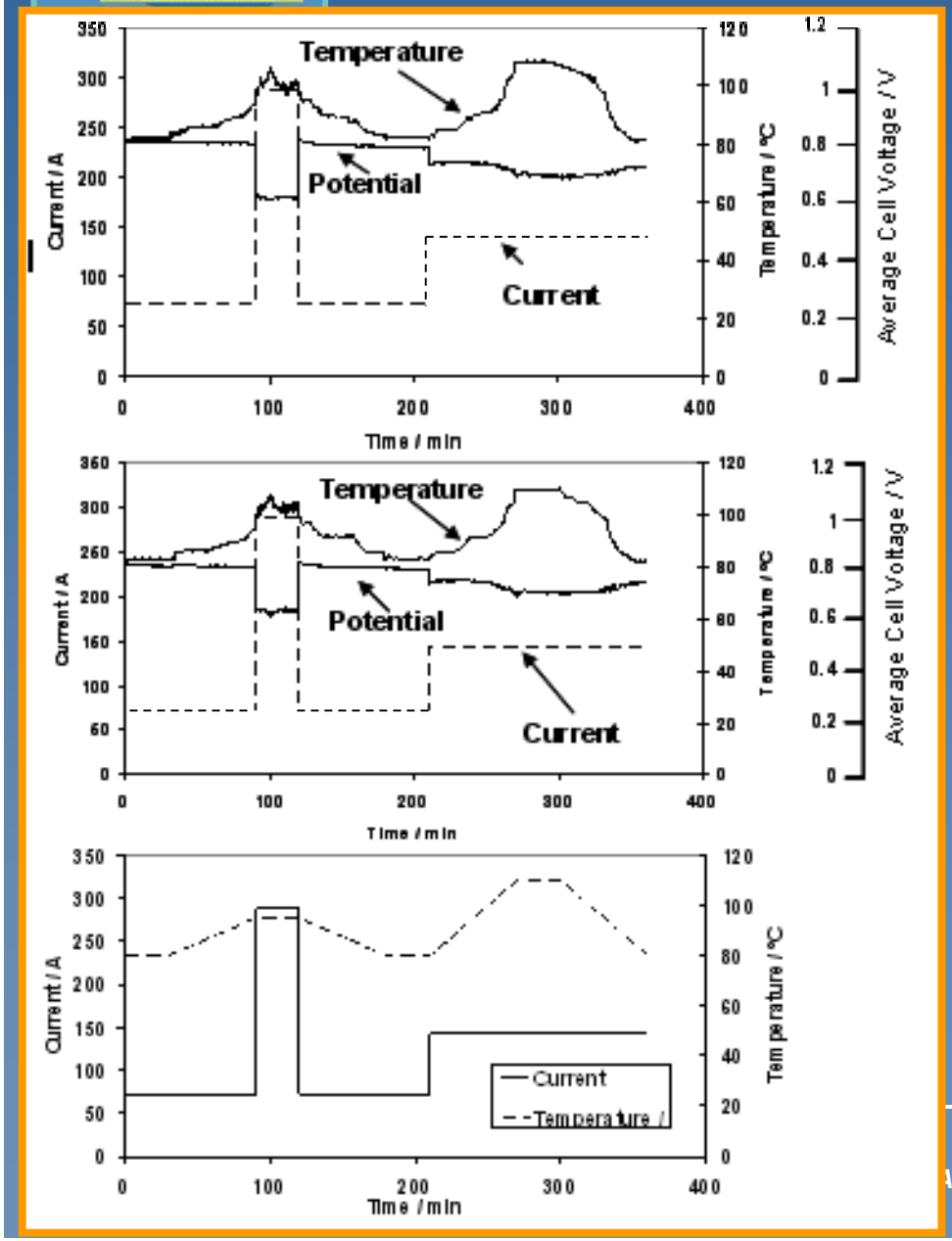


Operating Temperature/ Pressure
110 °C/ 1.5 bar abs./ 33 % R.H.

compression 13 kg cm^{-2}

Duty Cycles

JMFC MEAs-based stack; 2nd generation



Stack Compression 10 kg cm^{-2}

Duty cycles (top: initial; bottom final) with nominal temperature set point of the cooling apparatus. 5-cells short stack assembled with JMFC MEAs (active area 360 cm2).

Under practical operation conditions under automotive applications, the stack may experience rapid changes of current at both conventional and high working temperatures. Simplified duty cycles of current and temperature were thus applied to the stack and the corresponding variation of the stack voltage and actual temperature was monitored. The initial and final of several consecutive duty cycles are shown on the left.

Although some voltage/ temperature fluctuation was observed, no significant decay of performance was recorded after these cycles; the temperature deviation with respect to the set-point of the chiller was slightly larger than that observed in polarization experiments; however, the recorded stack voltage was similar to that observed in the polarization experiments for similar operating conditions and stack compression.

High Temperature Operation: Comments

- A power output larger than 1 kW was achieved up to a temperature of 100 °C with cells of 360 cm^2.

- The stacks showed a suitable power output up to 100 °C with power densities exceeding 600 mW cm^{-2} and a moderate decrease at slightly higher temperatures (110 °C). However, the performance decreased in the range 120°-130 °C possibly due to dehydration effects as a consequence of the decrease of relative humidity

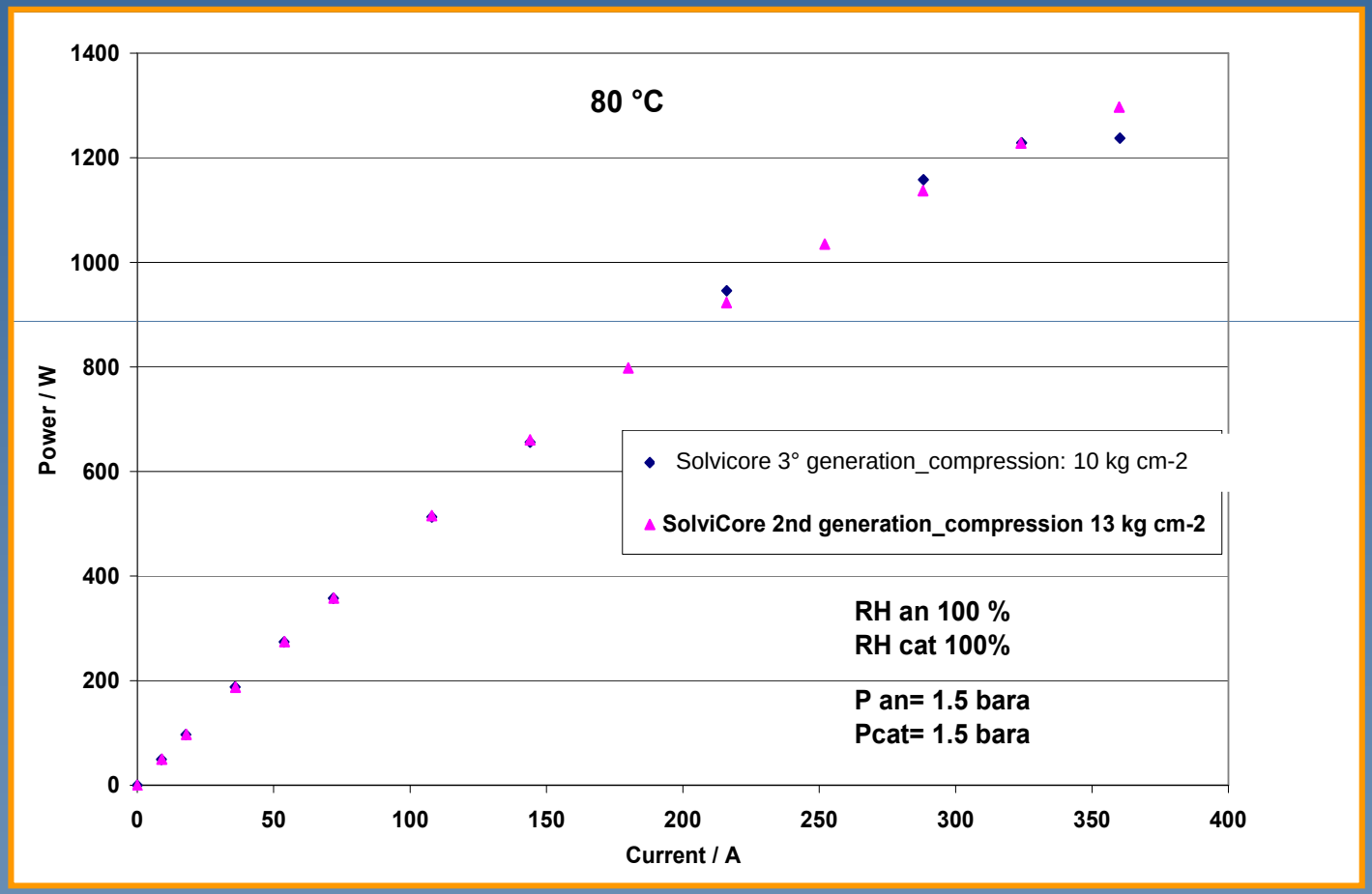
- The cell resistance increased from 0.05-0.08 to about 0.18-0.2 Ohm cm^2 passing from 80 °C to 110 °C, due to the membrane dehydration.

- Although the Aquivion membrane showed significantly better water retention properties at high temperature than conventional perfluorosulfonic membranes, the most effective proton conduction mechanism still relies on the so-called “vehicle mechanism” where proton transport is assisted by water molecules.

- The thin (30 μm) Aquivion membranes also favour the back-diffusion to the anode of the water produced at the cathode.
- Stack operation at high current densities is thus essential to promote the internal self-humidification of the MEAs.

Comparison between SolviCore 2nd and 3rd generation MEAs

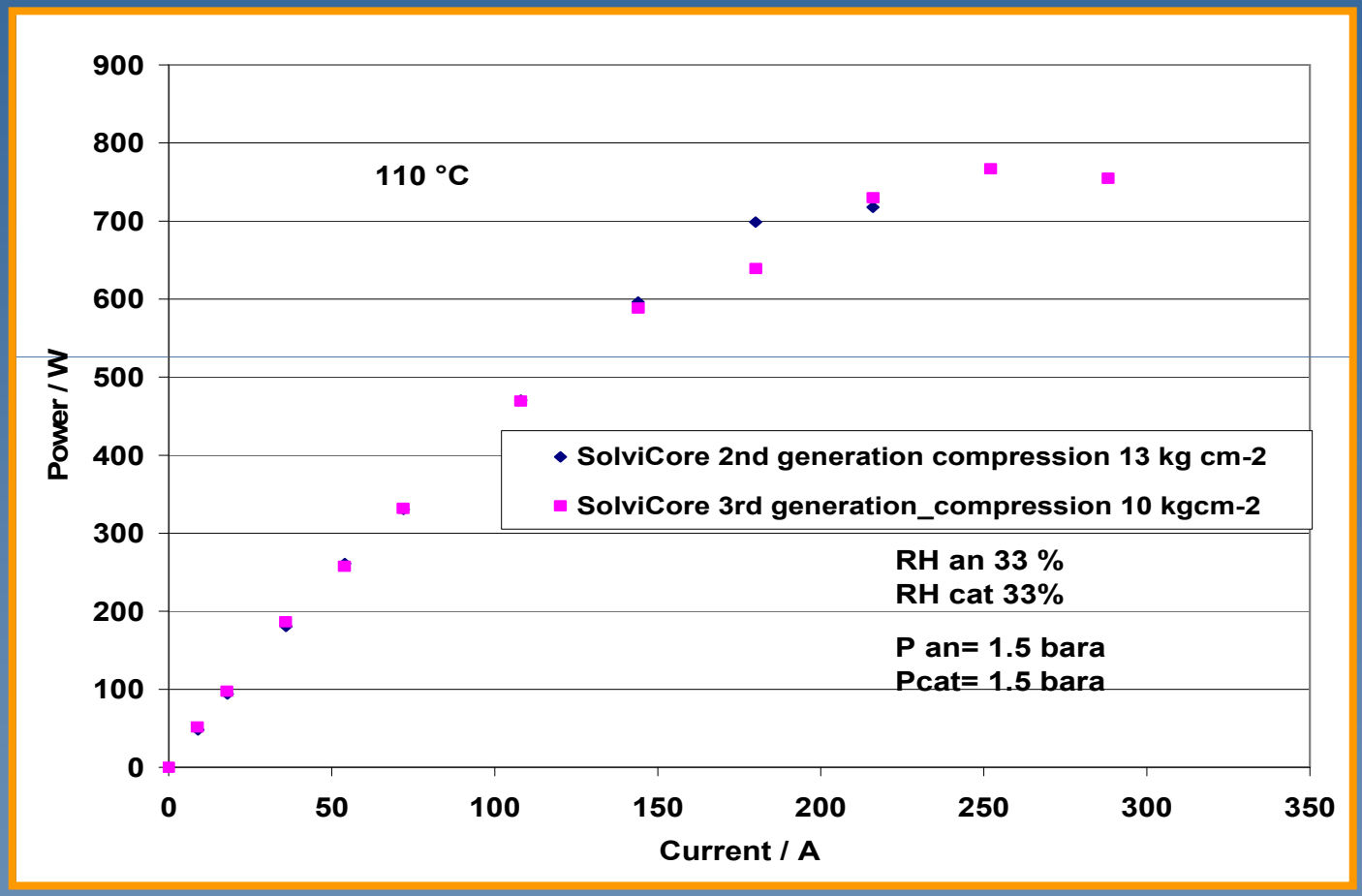
Measurements at **80 °C**



Same performance at lower compression for the third generation!

Comparison between SolviCore 2nd and 3rd generation MEAs

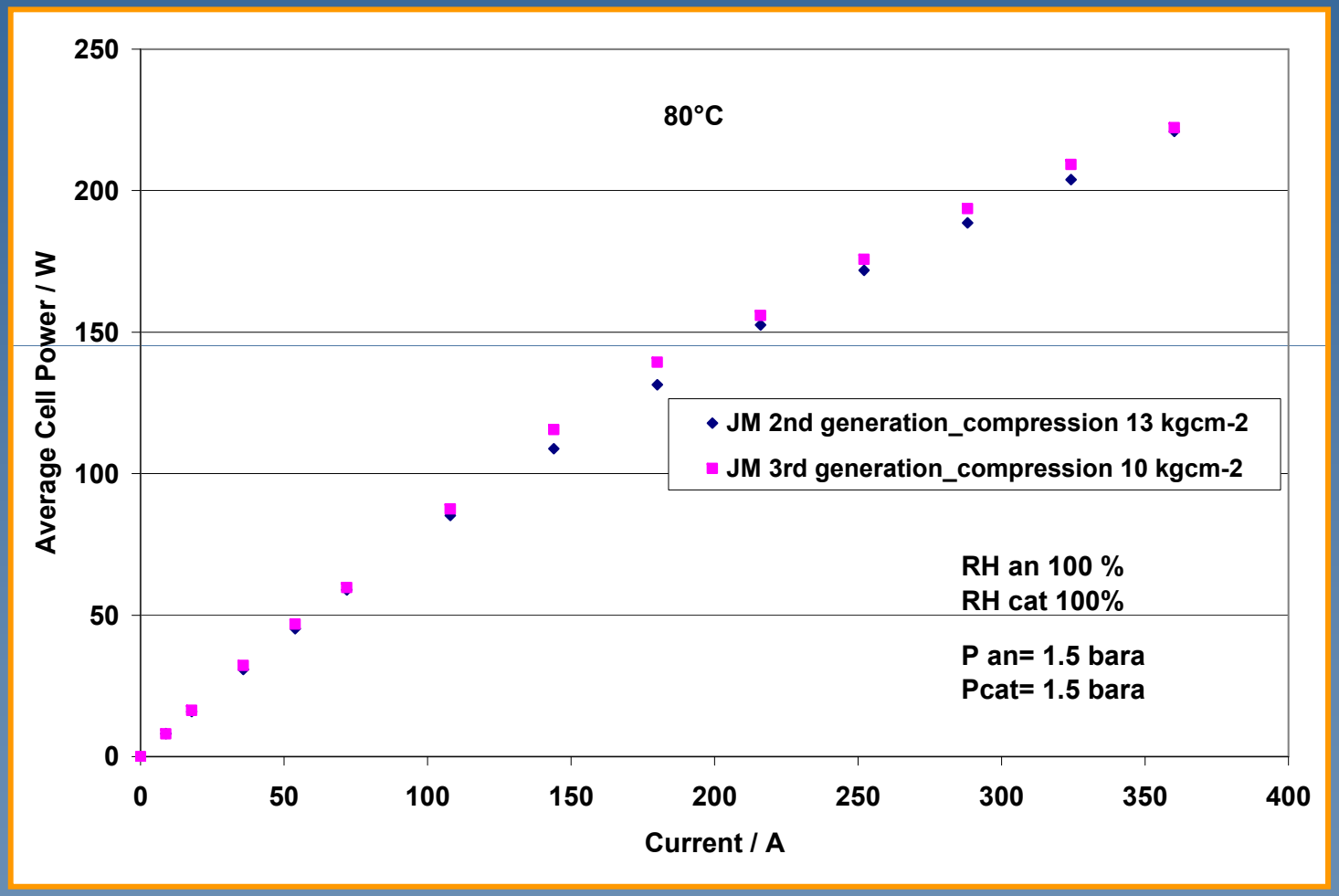
Measurements at **110 °C**



Same or better performance at low compression for the third generation!
The lower compression enhances stack reliability at high temperature

Comparison between JM 2nd and 3rd generation MEAs

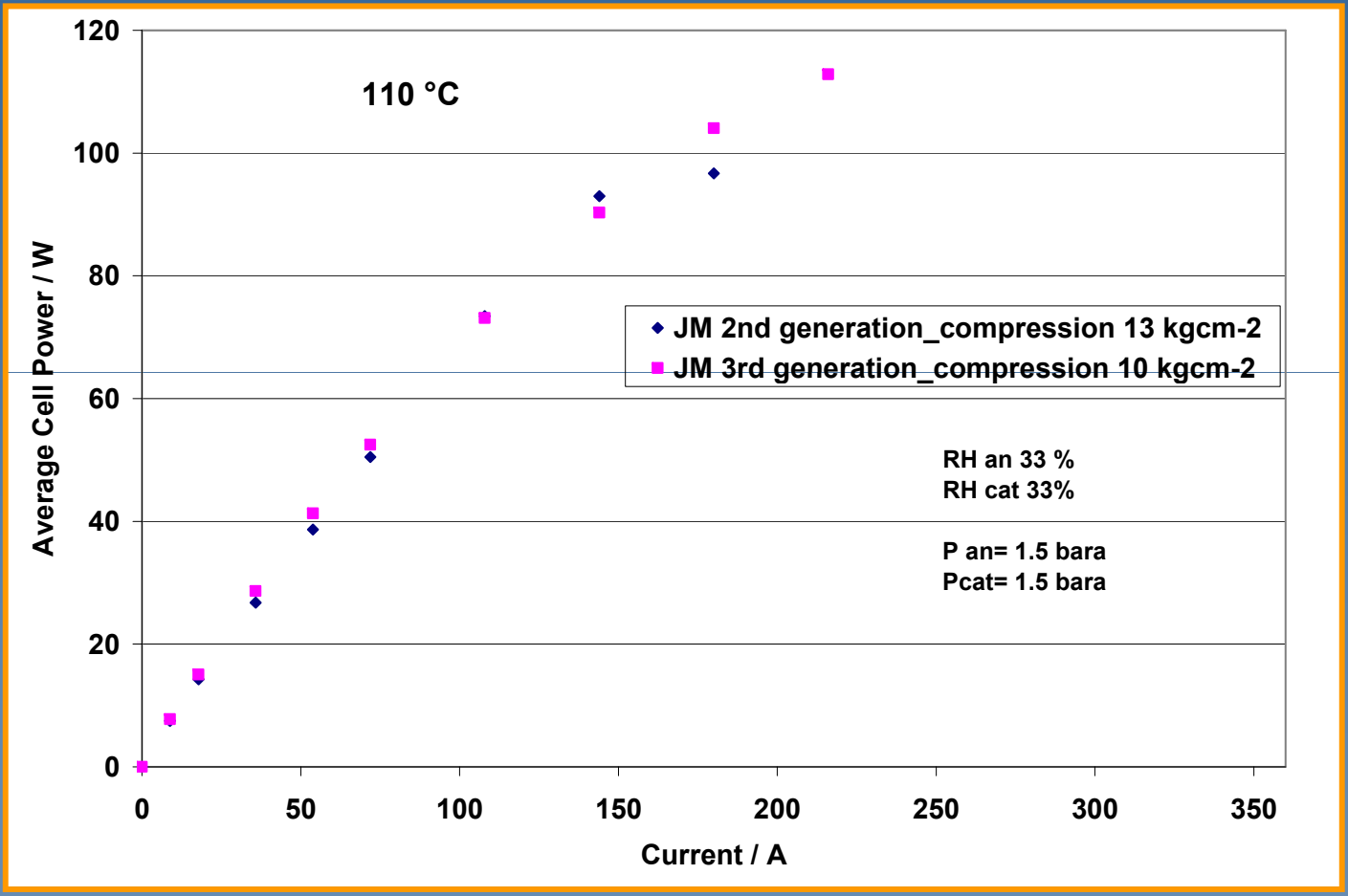
Measurements at conventional temperature (80 °C)



Same performance at lower compression for the third generation!

Comparison between JM 2nd and 3rd generation MEAs

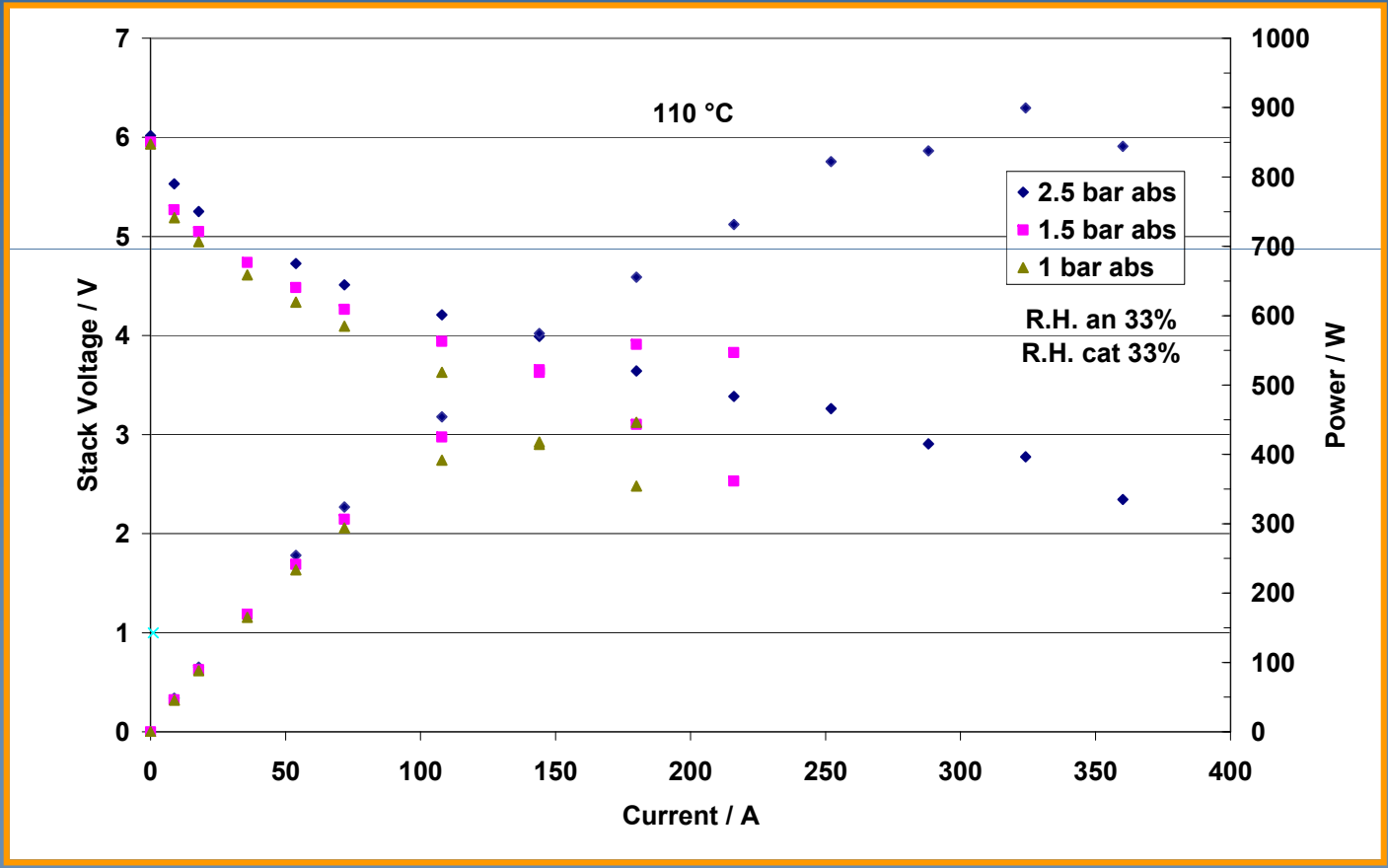
Measurements at 110 °C



Same or better performance at low compression for the third generation!
The lower compression enhances stack reliability at high temperature

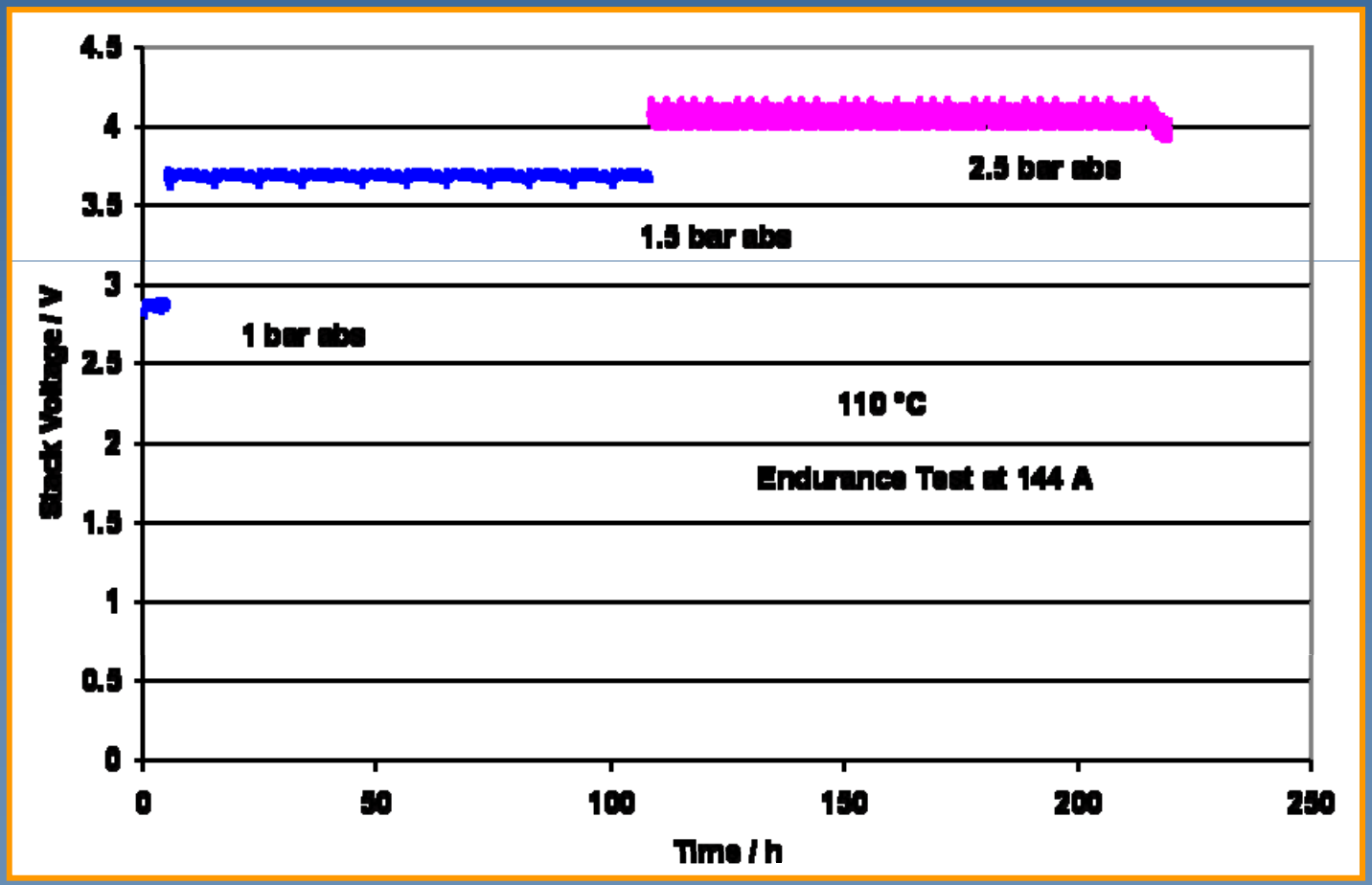
Stack Polarization and Power curves at different Pressures

Measurements at **110 °C**



Steady-state Galvanostatic Experiment with 3rd generation MEAs

110 °C, 144 A, 1-1.5 bar abs, 33 % R. H.



Comparison 3rd generation vs. 2nd generation MEAs

- For both the JMFC and Solvicore stacks the progress passing from the second generation to the third generation mainly consists on the fact that same or slightly better performance was achieved with lower compression.

- The lower compression enhances stack reliability at high temperature

CONCLUSIONS

• Electrochemical experiments in fuel cell short stacks were performed under automotive conditions at pressures of 1-1.5 bar abs. over a wide temperature range, up to 130 °C, with varying levels of humidity (down to 18% R.H.).

• The stacks based on large area (360 cm^2) MEAs showed elevated performance in the temperature range from ambient to 100 °C (cell power density in the range of 600-700 $mWcm^{-2}$) with a moderate decrease above 100 °C.

• The performances and electrical efficiencies achieved at 110 °C (cell power density of about 400 $mWcm^{-2}$ at an average cell voltage of about 0.6 V) are promising for automotive applications.

• The progress was registered passing from the second generation to the third generation: similar or slightly better performance was achieved with lower compression. This enhanced stack operation at high temperature.

• An output stack power approaching or better than 1 kW was achieved in a large temperature (30°C-110 °C) range with 6 cells-stack (360 cm^2 active area)

Acknowledgements

We thank our Coordinator Goesta Pfundtner and our colleagues from the other SPs for their contribution to this activity