Available online at www.sciencedirect.com
SCIENCE DIRECT®
Journal of Power Sources 145 (2005) 307–311
JOURNAL OF POWER SOURCES
www.elsevier.com/locate/jpowsour

Spatially resolved measurement of PEM fuel cells

Alex Hakenjos *, Christopher Hebling

Fraunhofer Institute for Solar Energy Systems (ISE), Heidenhofstr. 2, 79110 Freiburg, Germany

Accepted 28 January 2005
Available online 3 May 2005

Abstract

A measurement set-up is presented containing a multichannel frequency response analyser system together with a multichannel potentiostat that allows measurements of the local electrical impedance spectra of segmented fuel cells simultaneously.

For evaluation the current and impedance distribution is combined with optical spatially resolved measurement and numerical simulation.

Furthermore the multichannel frequency response analyser system is used for the measurement of impedance spectra of single cells in a fuel cell stack simultaneously.

Keywords: PEM; Fuel cell; Spatially resolved; Current distribution; Thermography; Impedance

1. Introduction

At Fraunhofer ISE, PEM fuel cell research focuses on the development of portable fuel cell systems [1]. Efficiency, energy density and reliability are the main characteristics to be optimised.

As most parameters that have a high influence on the different loss mechanisms (i.e. temperature, water content and gas concentration) are not distributed homogeneously over the active area of the fuel cell, measurements have to be carried out not only integrally but also spatially resolved.

One way to gather spatially resolved information of the performance and state of operation of a PEM fuel cell is the measurement of current distribution [2–5] and localised impedance spectroscopy [6]. In combination with this method, we use infrared thermography to record the temperature distribution on the active area.

With such measurements its possible to validate multidimensional models. PEM fuel cells are complex dynamic systems where the free flow of gases, diffusion in porous media, electrochemical reactions and the coexistence of liquid and gaseous phases interact. Therefore, a detailed insight into the state-of-operation and performance of the fuel cell can only be achieved by combining measurements with numerical simulation.

For whole fuel cell stacks, the simultaneous impedance spectroscopy of the single cells is capable of evaluating the state-of-operation of the single cells at the same time and therewith explaining overall stack performance.

2. Experimental

2.1. The measurement equipment

A test stand for spatially resolved measurement has been developed. Gas flow rates are controllable. Two bubblers humidify the anode and cathode gas stream. The gas humidity can be adjusted by mixing dry and humidified gas streams. The dew points of the anode and cathode gas streams are measured using dew point mirrors.

A Solartron 1254 frequency response analyser (FRA) with two 1251 multichannel extensions is available for simultaneous impedance measurement of up to 19 channels.

For stack measurements, two Kepco BOP 20/20 bipolar power supplies are available as load. They are operated in current control mode either in series for up to 40 V or in parallel

* Corresponding author.
E-mail address: hakenjos@ise.fhg.de (A. Hakenjos).

0378-7753/$ – see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.jpowsour.2005.01.075

for maximum currents of 40 A. For impedance measurement, they provide currents with an ac perturbation above a dc bias analog programmed by the generator output of the 1254 FRA.

For measuring current density and localised impedance, it is important to set all the segments to the same voltage. The diffusion layer and the electrode would allow lateral currents, if there are differences in the potential between adjacent segments. This disturbs the current measurement. A way to overcome this is to control the voltage of each segment separately with its own potentiostat.

A 50 channels multi-potentiostat was developed and is now available for spatially resolved measurements. Each channel sources our sinks a maximum current of 1 A and controls the voltage of one single segment separately. Current measurement is performed with shunt resistors in the current lines.

The multi-potentiostat is analog programmed with a common voltage. Superimposing an ac voltage to a dc bias at the programming voltage input enables spatially resolved impedance measurement.

As the measurement device is completely separated from the fuel cell different cells can be measured.

For further evaluation characteristic values can be extracted of the impedance spectra. In this paper the high frequency resistance is investigated, which denotes the absolute value of the impedance at 1 kHz. This is a reasonable value for the membrane resistance together with the backing, bipolar plate, and contact resistances [7,8]. Any change in this value during operation is assumed to be caused by a change in membrane ionic resistivity due to membrane hydration. Therefore the high frequency resistance is a measure for membrane water content.

2.2. Straight channel fuel cell

For studying the principles of fuel cell operation and for validation of simulations it is useful to reduce the flow field geometry to the simplest shape possible. This is the straight channel.

The cell used here was described previously [9], its geometry is shown in Fig. 1. To minimize boundary effects not only one but 14 parallel straight channels form the flow field on both the anode and cathode side. Each channel is 1 mm wide with 1 mm ribs in between. The channels are 100 mm long. This flow field geometry has an active area of 29 cm^2.

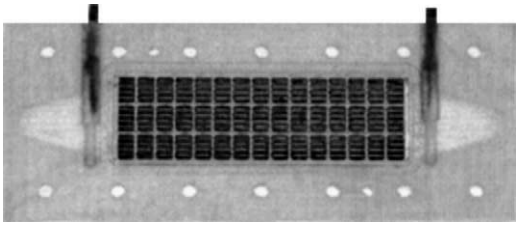

Fig. 1. The test fuel cell with straight channel design.

For current distribution measurement one flow field is segmented. Forty-five graphite segments have been integrated in an endplate made of plastic. The segments are arranged in three rows of 15. Each segment is connected with a separate current line and a voltage sense. The average of current distributions of the three rows of segments is used for further considerations.

The other side has water channels behind the flow field allowing to control the temperature of the fuel cell.

A Gore 5510 MEA with 0.4 mg platinum loading on both sides together with a Toray TGP-H-120 GDL is used.

The influence of the air flow on the current distribution was studied. Measurements with air flow rates of 600, 300 and 50 standard cubic centimeters per minute (sccm) are performed. The hydrogen flow rate is constant at 100 sccm dry hydrogen. Air is humidified to 80% at 22 °C. The cathodic flow field temperature is controlled to 55 °C. The measurements are performed at a cell voltage of 500 mV.

This measurements are compared with simulations [9] realised with a PEM model based on FLUENT^{TM} computational fluid dynamics (CFD) software [10].

Fig. 2 shows a comparison of the measured and calculated current distribution along the channel. A good agreement is found between measurement and simulation. Only the measured 50 sccm case shows a decline at segment positions three to eight that is not reproduced by the simulation.

Applying local impedance spectroscopy to the middle row of segments results in a row of impedance spectra shown in Fig. 3.

Here the high frequency resistance of the measurement is evaluated and compared to the simulated membrane protonic resistivity of the 300 sccm case, Fig. 4. Again a very good agreement is found. A high protonic resistance at the inlet that decreases along the channel due to increased membrane humidity is shown in both the measurement and the simulation.

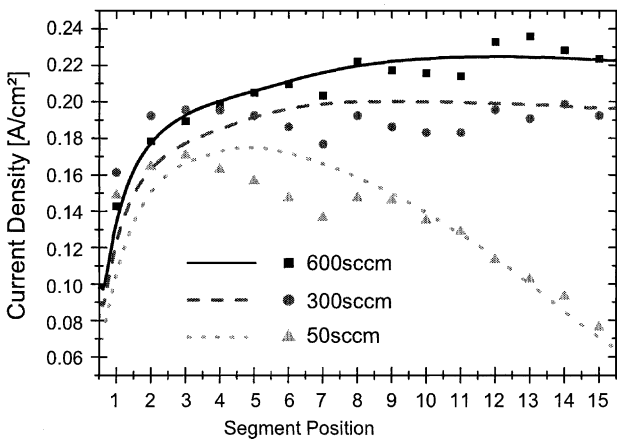

Fig. 2. Comparison of measured (··dots) and simulated (- - -lines) current distribution. Experimental parameters are: cell voltage 0.5 V; air humidity 80% at 22 °C; H_2 flow rate 100 sccm, dry; 55 °C cathode temperature. All model parameters are kept constant except air flow and average current density. Gases enter at position 1.

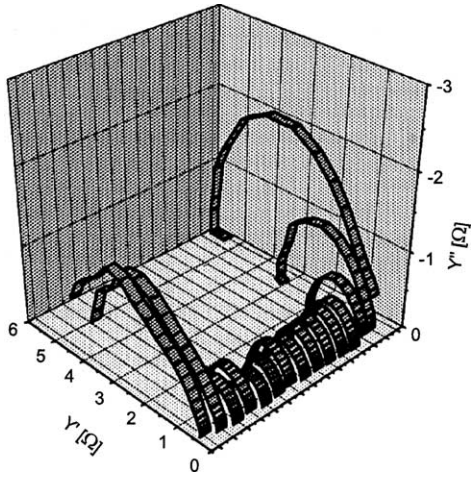

Fig. 3. Local impedance spectra of the middle row of the straight channel cell.

This validation of the fuel cell model encourages to trust in predictions made by the model, giving more insight into the characteristics of the local cell performance [9].

2.3. The square fuel cell

The square fuel cell shown in Fig. 5 allows the combination of several spatially resolved measurement methods [11]. Current density distribution, localised impedance, temperature distribution and condensed water can be observed simultaneously.

The flow field of the anode side is formed by square (2 mm × 2 mm) current collector areas divided by 1 mm wide gas channels. For current distribution and localised impedance measurement the flow field is segmented. Forty-nine stainless steel (1.4571) segments (5 mm × 5 mm) have been integrated into the endplate made of plastic. Each segment is connected with a separate current line and a voltage sense.

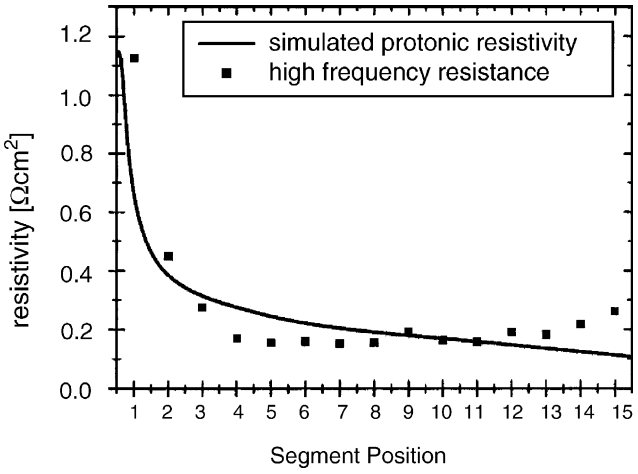

Fig. 4. The extracted high frequency resistivity of the impedance measurement shown in Fig. 3 in comparison with the simulated membrane resistivity of the 300 sccm simulation.

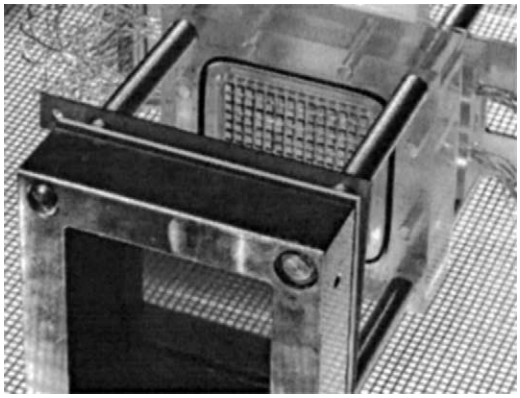

Fig. 5. The quadratic test fuel cell.

The gas channels of the cathode side are completely broken through to allow a straight view onto the diffusion layer. Behind the flow field a zinc selenide window closes the gas channels. As zinc selenide is transparent for infrared light, the temperature of the active area can be measured with an infrared camera.

The zinc selenide window is not only transparent for the IR but also for visible wavelength, it is possible to observe water droplets condensing in the gas channels.

A Gore 5510 MEA with 0.4 mg platinum loading on both sides together with a Toray TGP-H-120 GDL is used.

Fig. 6 shows the serpentine air flow field used here as cathode flow field. The flow direction is indicated. Hydrogen flow direction is from top to bottom.

To measure the impedance distribution the ac currents of 16 segments are analysed with the FRA. In Fig. 6, the segments chosen for impedance measurement are shaded. In combination with the ac voltage, the impedance spectra are calculated.

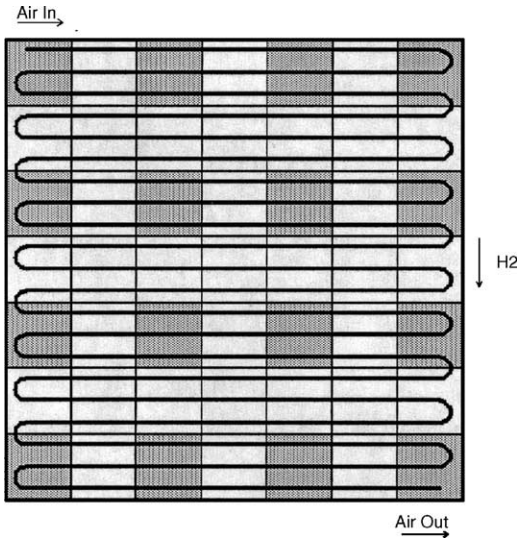

Fig. 6. The cathode flow field and the position of the segments of the square fuel cell.

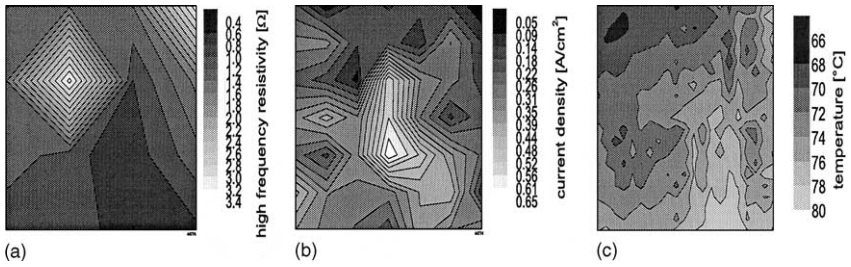

Fig. 7. (a) High frequency resistance, (b) current density and (c) temperature distribution of the square fuel cell.

The quadratic cell was operated with gas flow rates of 500 ml min^{-1} air and 300 ml min^{-1} hydrogen. The cell voltage was 600 mV. The gases were humidified to a dew point temperature of 25 °C and entered the cell at a temperature of 50 °C. Fig. 7 shows the high frequency resistance, current density, and temperature distribution after the cell settled to a fairly stable state of operation.

The high frequency resistivity distribution shows one spot with high conductivity. This corresponds to the current distribution showing its maximum in this region. The high resistivity of the upper right corner and the upper left lead to a low current density in these areas. The temperature is mainly determined by the local activity, whereas high currents produce more heat. This effect is superimposed by the cooling of the gas streams entering relatively cold at the top left and leaving the cell at the bottom right (air) or entering at the top and leaving at the bottom (hydrogen), respectively.

2.4. A spatially resolved stack measurement

Looking at a fuel cell stack, things become even more complicated. In this section, the coordinate of spatially resolution is perpendicular to the active area. It is along different cells of a fuel cell stack. Not only the cell voltages but also the impedances are measured during operation [12].

A self-developed short stack consisting of four cells with an active area of 53 cm^2 each is observed. The stack was operated with a current of 4 A. This is a realistic partial load point for a fuel cell stack in a system during standby operation. The voltages of the individual cells are shown in Fig. 8.

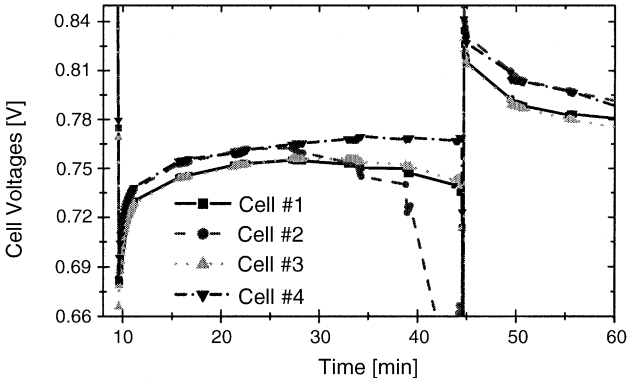

Fig. 8. The voltages of the individual cells.

The load is switched on 9.5 min after applying the reactant gases. The gas flow rates are set to 300 ml min^{-1} dry hydrogen for the anode and 1000 ml min^{-1} for the cathode.

Impedance spectra are taken approximately every 7 min. The duration of the measurement of one spectrum is a ≈4.5 min. During the impedance measurements the cell voltages are not monitored.

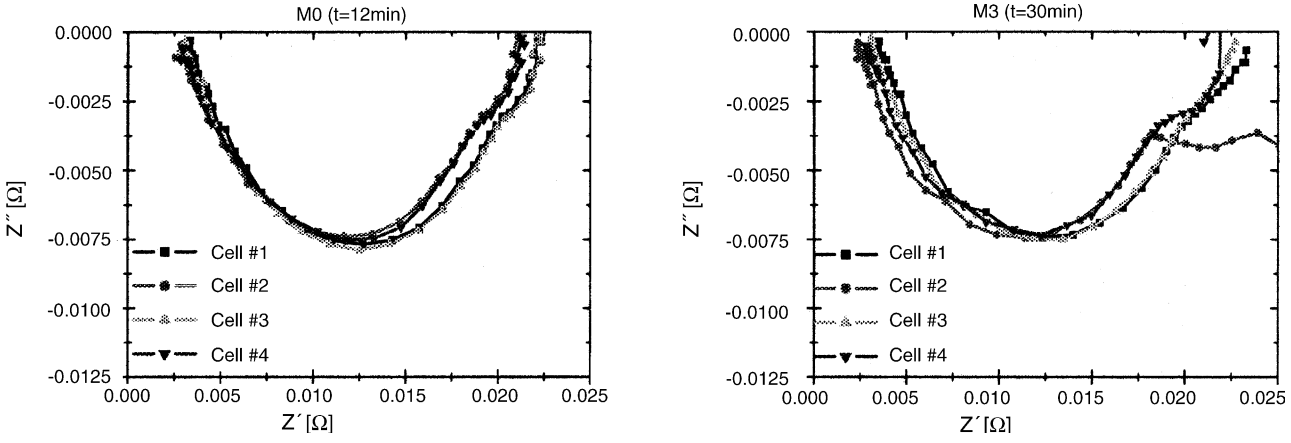

Fig. 9. The impedance spectra measured at different times after start of experiment.

The impedance measurement after 30 min Fig. 9-M3 shows an increase of the low-frequency arc in the spectrum of cell 2. This indicates an increase in diffusive losses due to flooding of the cell. The cell voltage decreases to the level of cells 1 and 3.

The voltage of this cell drops dramatically 9 min after the first evidence in the impedance spectrum, Fig. 8 (39 min). To overcome this performance loss, the cathode outlet of the stack is closed momentarily to apply a pressure release impulse to the cathodic gas stream. This removes the liquid water off the gas channels. Afterwards, the stack performance returns to even higher values than before.

3. Conclusion

Spatially resolved current density measurement is necessary for fuel cell characterisation. Segmented fuel cells can provide much more information about the state of operation than integral measurements.

With the system presented current density, electrical impedance, temperature distribution, and condensed water can be observed simultaneously in a fuel cell under operation.

Spatially resolved measurements on different fuel cells are described. The combined information of the different spatially resolved measurements enables a deeper insight into fuel cell state of operation. In segmented cells, areas of low current density and the associated loss mechanisms can be identified. Data for the validation of fuel cell models can be provided, since for the validation of multidimensional models, polarisation curves are not sufficient. The used model shows a good agreement with the measurements of local current density and high frequency resistance.

The simultaneous impedance measurement of the individual fuel cells in a stack offers a more detailed insight into the fuel cell performance than is possible with integral measurement. A flooding event in a single cell is observed with the impedance measurement minutes before it shows up as a voltage drop.

Acknowledgement

This work is sponsored by the scholarship programme of the German Federal Environmental Foundation DBU.

References

[1] K. Tüber, M. Zobel, H. Schmidt, C. Hebling, A polymer electrolyte membrane fuel cell system for powering portable computers, J. Power Sources 122 (2003) 1–8.
[2] S.J.C. Cleghorn, C.R. Derouin, M.S. Wilson, S. Gottesfeld, A printed circuit board approach to measuring current distribution in a fuel cell, J. Appl. Electrochem. 28 (1998) 663–672.
[3] Ch. Wieser, A. Helmbold, E. Gülzow, A new technique for two-dimensional current distribution measurements in electrochemical cells, J. Appl. Electrochem. 30 (2000) 803–807.
[4] M. Noponen, T. Mennola, M. Mikkola, T. Hottinen, P. Lund, Measurement of current distribution in a free-breathing pemfc, J. Power Sources 106 (2002) 304–312.
[5] D.J.L. Brett, S. Atkins, N.P. Brandon, V. Vesovic, N. Vasileiadis, A.R. Kucernak, Measurement of the current distribution along a single flow channel of a solid polymer fuel cell, Electrochem. Commun. (2001) 628–632.
[6] D.J.L. Brett, S. Atkins, N.P. Brandon, V. Vesovic, N. Vasileiadis, A.R. Kucernak, Localized impedance mesurements along a single channel of a solid polymer fuel cell, Electrochem. Solid-State Lett. 6 (2003) A63–A66.
[7] A. Parthasarathy, B. Dave, S. Sirinivasan, A.J. Appleby, The platinum microelectrode/nafion interface: an electrochemical impedance spectroscopic analysis of oxygen reduction kinetics and nafion characteristics, J. Electrochem. Soc. 139 (6) (1992) 1634–1641.
[8] P. Kurzweil, H.-J. Fischle, A new monitoring method for electrochemical aggregates by impedance spectroscopy, J. Power Sources 127 (2004) 331–340.
[9] A. Hakenjos, K. Tüber, J.O. Schumacher, C. Hebling, Characterising PEM fuel cell performance using a current distribution measurement in comparison with a CFD model, Fuel Cells 3 (2004) 185–189.
[10] M.T. Prinkey, M. Shahnam, W.A. Rogers, R.S. Gemmen, Validation and application of a CFD-based PEM fuel cell model, in: Proceedings of the Fuel Cell Seminar, Palm Springs, 2002.
[11] A. Hakenjos, H. Muenter, U. Wittstadt, C. Hebling, A PEM fuel cell for combined measurement of current and temperature distribution, and flow field flooding, J. Power Sources 131 (2004) 213–216.
[12] A. Hakenjos, M. Zobel, J. Clausnitzer, C. Hebling, Simultaneous electrical impedance spectroscopy of single cells in a PEM fuel cell stack, in: Ulm Electrochemical Talks 2004, J. Power Sources, submitted for publication.