Available online at www.sciencedirect.com
SCIENCE DIRECT®
Journal of Power Sources 145 (2005) 62–67
JOURNAL OF POWER SOURCES
www.elsevier.com/locate/jpowsour

Short communication

Dependence of current distribution on water management in PEFC of technical size

Felix N. Büchi [a,*], Arne B. Geiger [a], Rui P. Neto [b,1]

[a] *Paul Scherrer Institut, Electrochemistry Laboratory, CH-5232 Villigen PSI, Switzerland*
[b] *Instituto Superior Técnico, Av. Rovisco Pais, 1049-001 Lisboa, Portugal*

Received 6 August 2004; accepted 31 December 2004
Available online 19 February 2005

Abstract

Measuring local currents in PE-fuel cells is an important tool for diagnostics and development. A semi-segmented cell has been developed, which can serve as a key instrument to investigate different phenomena in cells and stacks of technical relevance. Data with respect to water management is presented. These results show, that the local current distribution is strongly influenced by the dew point of the process air, the stoichiometry of the process air and the mode of operation.

Keywords: Polymer electrolyte fuel cells; Local current measurements; Water management; Humidification

1. Introduction

The performance of a PEM-fuel cell depends on many interdependent parameters. These are on the one hand the properties of the electrochemical components such as activity of the electro-catalyst, transport properties of the electrodes (transport of gases and liquid water through the porous structures) and conductivity of the membrane. On the other hand the properties of the bipolar plate with respect to distribution of the gases over the active area, heat dissipation properties and electric conductivity in the bipolar arrangement influence the performance. Finally also the operation parameters such as gas pressures, gas stoichiometries, gas dew points and cell temperatures are important for the overall performance. A number of the parameters are interlinked, and especially the water management depends on several parameters such as water sorption and transport properties of the membrane and the electrodes, transport of water in the flow field, the cell temperature, gas dew points, gas stoichiometries and global as well as local current density.

In a standard fuel cell experiment changing voltage or changing average current density of the cell is the only measurable response on variation of any of the parameters influencing the water management. The interpretation of the measurement with regard to optimization of the cell performance remains difficult. This is because the reasons for the change in cell performance are not easily deductible in a straight forward manner.

There are several options to gain more information from such experiments. One of these is measuring the current density locally resolved at different locations of the active area of the cell. Knowledge of the current distribution over the active area of the cell can provide useful information on the underlying mechanisms and can therefore be used for optimizing components or as validating input to support modeling efforts.

For local current density measurements, in principle, two different approaches are feasible: (i) construction of a model-cell, optimized for local current density measurements with respect to accuracy and resolution [1,2]; (ii) adopting a real

* Corresponding author. Tel.: +41 56 3102411; fax: +41 56 3104415.
E-mail addresses: felix.buechi@psi.ch (F.N. Büchi), rn@sre-fc.com (R.P. Neto).
[1] Present address: Soluções Racionais de Energia, 2565-641 Ramalhal Torres Vedras, Portugal.

0378-7753/$ – see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.jpowsour.2004.12.039

cell with the necessary instrumentation [3]. In the first case accurate measurements are possible, but these results would be difficult to be transposed to cells of technical importance, mainly due to scaling factors and thermal management differences. In the second case the challenge is, to include the instrumentation for locally resolved current measurement in the cell without making changes to the gas flow field to conserve fluid dynamic properties, and to the thermal properties of the cell (cooling, temperature profiles). In this work, we pursued the approach to use a technical cell (developed for an automotive power train [4,5]) in order to gain information on the "real" system. The construction described in detail below would be suitable to be included in a stack. The results presented however, are measured in a single cell.

2. Semi-segmented plate principle

The principle of adopting a real cell for the local current density measurements was chosen, because inclusion of the measurement principle into a big stack should be possible. For this purpose a "semi-segmented" cell, including an unchanged air flow field plate, as used in the stacks [4,5], was developed which allows for local current density measurements but conserves the fluid dynamic properties, as well as the electrical, thermal, and mechanical properties exactly equivalent to those of a terminal plate in a stack. Therefore, the semi-segmented part of the cell may also be included as endplate in a stack. The semi-segmented endplate is shown schematically in Figs. 1 and 2. It is based on the idea, that the highly conducting part of the plate, made from sintered graphite ($\rho \approx 1\,\mathrm{m\Omega\,cm}$, type NS2, SGL Carbon) is completely segmented after being glued to the flow field plate. This thin flow field plate, which is pressure molded from a graphite polymer mixture [4], is equivalent to plates in a stack and has a comparably high specific resistivity ($\rho \approx 25\,\mathrm{m\Omega\,cm}$, type BMA5, SGL Carbon). But this plate remains intact and guarantees for unchanged electrochemical, fluid dynamics and thermal properties, as well as the compatibility with the stack (see Fig. 1). While for standard cells the comparably high resistivity of the molded plate is considered a disadvantage because it results in a small voltage loss, and therefore

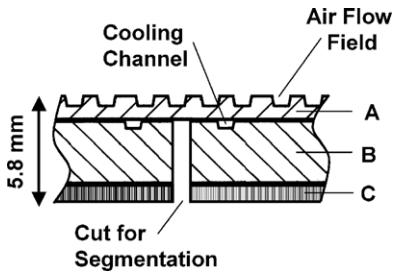

Fig. 1. Schematic of semi-segmented plate: (A) molded low conductivity plate with flow field; (B) high conductivity graphite plate; (C) metal current collector.

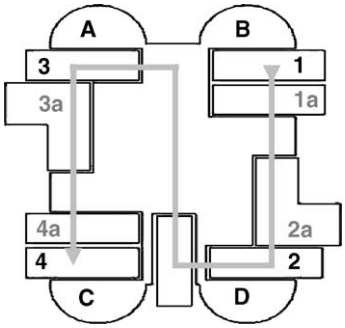

Fig. 2. Arrangement of segments in semi-segmented plate. Arrow indicates schematic path of air flow in the plate, flowing from manifolds B to C. Hydrogen flows in a similar manner from A to D.

slightly reduces the performance of the stack, in case of the semi-segmented plate it is an advantage because it limits the current crossing to neighboring segments. This effect is increased by the low thickness of the molded flow field plate, which has a maximum thickness of 1.5 mm, but an average thickness in the flow field of less than 1 mm. The resulting resistance between two adjoining segments is $0.5 - 1\,\mathrm{m\Omega}$. This value seems low, but it is big enough to prevent substantial communicating currents between individual segments when the used external circuit is tuned to even lower differences in resistance.

The semi-segmented cathode flow field plate was completed with a standard anode (hydrogen flow field). The total active area of the cell is $200\,\mathrm{cm^2}$. The location of the individual segments was chosen to cover the parts of the active area where highest inhomogenieties of the current density are expected, namely the locations of air and hydrogen in- and outlets. These are in the corners of the flow field. The active area was split up in 10 segments, 9 small segments of $10\,\mathrm{cm^2}$ and a big central segment of $110\,\mathrm{cm^2}$ (see Fig. 2). In Fig. 2, all 10 segments for which the current was recorded individually are shown. In the results section only data from segments 1–4 are discussed, because the current density in segments from 1a to 4a is similar to the neighboring segments 1–4, and therefore does not provide substantial additional information.

3. Experimental

Nafion 112® (Dupont) and standard Elat (E-Tek Inc.) electrodes ($1\,\mathrm{mg\,Pt\,cm^{-2}}$ MEA) were used as electrochemical components. The single cell was assembled with a standard anode flow field plate and the semi-segmented cathode flow field plate.

The cell measurements were made in a microprocessor controlled test station, which accurately controls gas flows, gas pressures, cell current or cell voltage. The cell is water cooled, with an integrated cooling flow field on anode and cathode side. An enhanced cooling water flow (temperature difference between inlet and outlet $<0.5\,^{\circ}\mathrm{C}$) allows for an almost perfectly isothermal operation of the cell.

If not otherwise mentioned the cell was operated under the following standard conditions of 70 °C cell temperature, gas pressures of 2 bar (abs), hydrogen stoichiometry of $\lambda_{H_2} = 2$ and a hydrogen dew point of 70 °C. The measurements reported are steady state data, recorded after at least 30 min of previous constant operation conditions.

The individual segments on the cathode side of the cell were connected to separate current leads. The current in these leads was measured by hall sensors and recorded via standard ADC equipment in a separate computer. After the current measurement instrumentation the individual current leads were united and connected to the load box. The resistance of the connections between the cell and the point of joining was very carefully calibrated by adjusting the length of the cables, in order to achieve a resulting difference in resistance of less than 0.1 mΩ. Individual currents were sampled (0.5 Hz) and averaged over 5 min to eliminate statistical fluctuations.

4. Results

Single cells can be operated in different regimes with respect to electrical and gas flow conditions. Constant current (CC) or constant voltage (CV) modes are applicable as the electrical regimes and constant flow (CF) or stoichiometric flow (SF) as the mass flow regimes. Three, out of the four possible combinations, have been investigated. The CC/SF regime is the one seen by a cell in a stack, and therefore of practical interest, the modes CV/SF and CC/CF are of academic interest in order to learn about the effects of changes of operating parameters on the current density distribution cell.

In the PE-fuel cell technology a number of operating parameters influence the current density distribution in the cell. Among the most important parameters are the stoichiometry and the dew point of the process air. These two parameters are also interesting from the system developers point of view, because the process air compressor, and process air humidification are important issues for the fuel cell system optimization.

The constant current and stoichiometric gas flow mode (CC/SF) is of highest practical interest, because an individual cell in a stack sees always a constant current at a certain stoichiometry independent of whether the stack itself is operated in controlled power, current or voltage mode. Constant current mode means that the cell, independent of its state, must carry a given total current. If in one location, i.e. due to drying, the current density is reduced, and then another segment of the cell has to produce more current.

If the cell is operated under standard ($T_{cell} = 70$ °C, p(gases) = 2 bar) and well-humidified conditions ($DP_{air} = 70$ °C), then the current density distribution in the cell is very homogeneous for $\lambda_{air} > 1.5$ (see Fig. 3). If the stoichiometry is lowered, then the segments at the end of the air path start to produce less current due to oxygen depletion, and consequently the segments early in the air path have to produce more current. At $\lambda_{air} = 1.1$ the current density in the first segment (segment 1) is almost three times higher than in the last segment (segment 4).

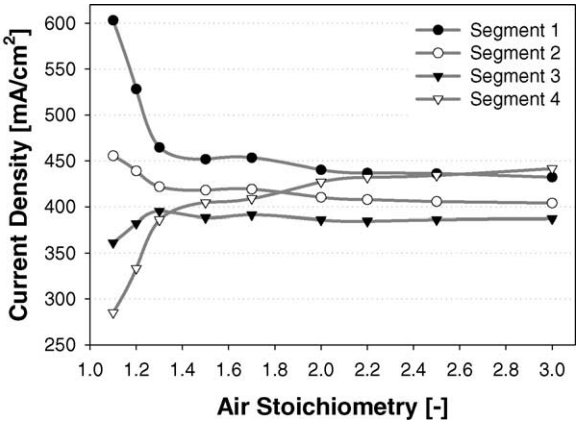

Fig. 3. Current density in the segments 1–4 as function of air stoichiometry. Average current density in entire cell 400 mA cm^{-2}. Dew point of air 70 °C, otherwise standard conditions.

It must be noted here, that stoichiometry-values are always given for the entire cell and not for individual segments. For example, segment 1 at the air-inlet sees a local stoichiometry of $\lambda_{air} = 30$ (for a cell-stoichiometry of $\lambda_{air} = 1.5$), because the entire mass flow for the complete cell passes over it's relatively small area.

This is important if the cell is operated with air of a reduced dew point of 30 °C (see Fig. 4). Still, the oxygen depletion effect is observed at the low stoichiometries, but at high λ_{air}, the order of the segments with respect to current density is reversed. The last segment in the air path (4) carries the highest current. This is due to the drying of the membrane (and increasing its resistance) early in the air path due to the passage of the high mass flow of comparably dry air. The dew point of the air increases along the path due to the product water and therefore current density increases as well.

In constant voltage and controlled gas stoichiometry mode (CV/SF, cell voltage 0.65 V, standard conditions with respect

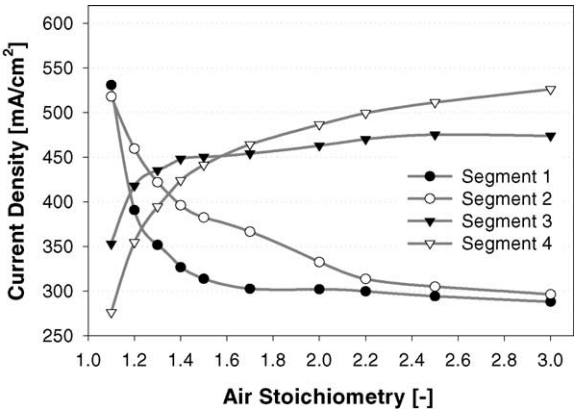

Fig. 4. Current density in the segments 1–4 as function of air stoichiometry. Average current density in entire cell 400 mA cm^{-2}. Dew point of air 30 °C, otherwise standard conditions.

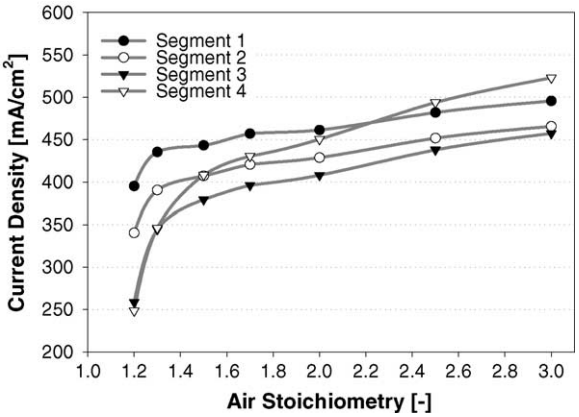

Fig. 5. Current density in the segments 1–4 as function of air stoichiometry. Cell operated in constant voltage mode at 0.65 V at standard conditions. Dew point of air 70 °C.

to gas pressures and stoichiometries) the total cell current, and with this the current distribution in the cell, is dependent on the operating conditions. In Figs. 5 and 6, the local currents for the four corner segments in the plate are shown as function of the air stoichiometry for a high and a low air dew point. It can be seen from Fig. 5 (DP_{air} = 70 °C), that the current density is increasing with increasing stoichiometry in all segments. For the low air stoichiometries the current densities of the segments closer to the exit (3 and 4) are lower than the current densities at the segments near the inlet (1 and 2). This is due to the higher relative decrease in oxygen concentration at the lower stoichiometries. If the cell is operated with air with a dew point much lower than the cell temperature of 70 °C, then the picture changes. Now the segment closest to the air-inlet always produces the lowest current density, because of membrane drying This is also observed at low stoichiometries (λ_{air}, 1.3–1.5) when oxygen concentration at the exit-segments is considerably lower, indicating that the oxygen concentration effect is overridden by the humidification effects (the air is gradually humidified along the flow

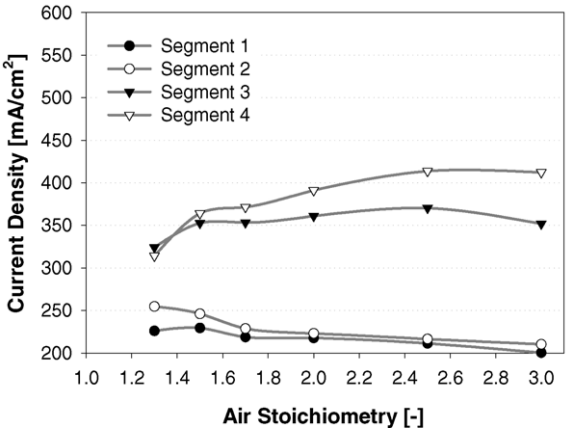

Fig. 6. Current density in the segments 1–4 as function of air stoichiometry. Cell operated in constant voltage mode at 0.65 V at standard conditions. Dew point of air 30 °C.

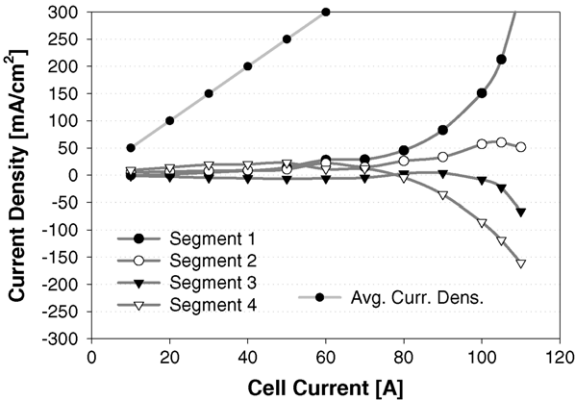

Fig. 7. Deviation of current density from average current density in the segments 1–4 as function of total cell current. Cell operated in constant flow mode with gas flows rates corresponding to λ_{H_2} = 2 and λ_{air} = 1.5@80 A total cell current (400 mA/cm²). Dew point of air 70 °C, otherwise standard conditions.

path). At the very low stoichiometries of λ_{air} < 1.3 where the oxygen concentration effect would be expected to become dominant, the cell could not be operated in a stable way. At the higher stoichiometries of λ_{air} > 1.5 the drying effect at the segments early in the air path is even more distinct and the segments closer to the exit clearly produce more current.

In the constant flow mode (CC/CF), the varying parameter is the total cell current. The gas flows are held constant at a flow rate corresponding to stoichiometries of λ_{air} = 1.5 at 80 A and λ_{H_2} = 2.0 at 80 A. Eighty amperes corresponds to an average current density of 400 mA cm^{-2}.

With the variation of the total current not the absolute current density, but the deviation from the average current density becomes of interest. In the case of well-humidified air (DP_{air} = 70 °C, Fig. 7), the deviations from the average current density are small for currents <80 A. At currents >80 A the oxygen concentration is reduced at the outlet segments due to consumption and the oxygen concentration effect becomes clearly visible. When feeding air with a low dew point (DP_{air} = 30 °C, Fig. 8) then the effect of drying of the MEA is visible for the early segments. For currents up to 60 A, the deviation from the average current density is approximately proportional to the current, indicating an increased resistance at the first two segments due to drying. At currents >80 A again the oxygen depletion becomes the dominant effect, as for the previous operating conditions with stoichiometric flow regime.

In all three cases of mode of operation (constant voltage, constant current and constant flow) discussed here, only the results for the extreme cases of air humidification (dew points close to cell temperature, and near room temperature) have been discussed. In all three cases the current density distribution changes gradually from the one to the other extreme when the air dew point is changed gradually.

An important parameter for the water management in PE-fuel cells is the cell temperature, because water vapor pressure, and therefore drying and flooding phenomena, is a

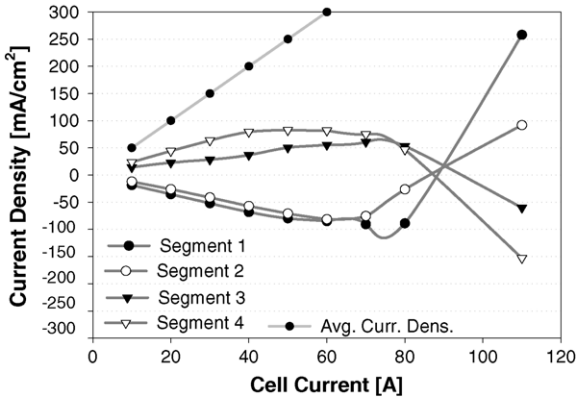

Fig. 8. Deviation of current density from average current density in the segments 1-4 as function of total cell current. Cell operated in constant flow mode with gas flows rates corresponding to $\lambda_{H_2} = 2$ and $\lambda_{air} = 1.5$ at 80 A total cell current (400 mA/cm^2). Dew point of air 30 °C, otherwise standard conditions.

strong function of temperature. Fig. 9 shows the dependence of the local current densities, in CC/SF mode at a cell temperature of 50 °C and low air humidity ($DP_{air} = 30$ °C). Except for the cell temperature, these are the same conditions as in Fig. 4. Comparing the two results shows that with the lower cell temperature (Fig. 9) segments 1 and 2 carry higher currents, indicating, that the effect of drying in the beginning of the air path is less at the lower cell temperature.

However, for many applications an operating temperature, as high as possible, and process air humidification as low as possible is dictated by the balance of plant. In mobile applications this reduces the size of the heat exchanger, and in stationary applications it enables easier use of the waste heat. Therefore the effects of uneven current distribution will generally have to be dealt with in real world applications.

Fig. 10 shows the dependence of the local current densities in CC/SF mode on hydrogen stoichiometry under standard conditions at 50 °C cell temperature. The influence of the hydrogen stoichiometry on the current density distribution is modest. Only at the very lowest hydrogen stoichiometry a

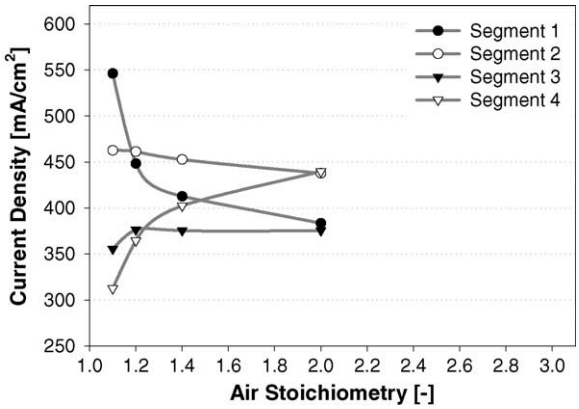

Fig. 9. Current density in the segments 1–4 as function of air stoichiometry. Average current density in entire cell 400 mA cm^{-2}. Cell temperature 50 °C, dew point of air 30 °C, otherwise standard conditions.

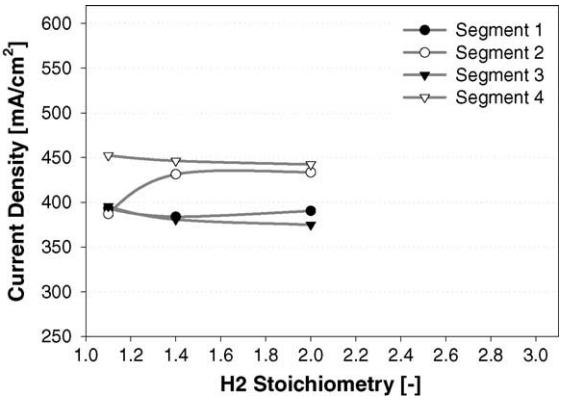

Fig. 10. Current density in the segments 1–4 as function of hydrogen stoichiometry. Average current density in entire cell 400 mA cm^{-2}. Cell temperature 50 °C, dew point of air 30 °C, otherwise standard conditions.

small decrease of current density at segment 2 is observed. Segment 2 is located at the exit of the excess hydrogen from the cell. A decrease at this segment can probably be attributed to accumulation of water in this location, because the velocity of hydrogen flow at the low stoichiometries is low, close to the exit.

Generally, under the conditions investigated, the influence of hydrogen stoichiometry on the current density distribution is much less than the influence of the air stoichiometry. This is because pure hydrogen is used, and therefore no concentration gradients evolve along the gas channel. Further, the volume flow at the anode is 2.5 times less at same stoichiometry, than on the cathode. This means, that also the amount of water vapor, which can be transported into the cell, or removed from the cell, is less on the anode side than on the cathode side. Therefore, the influence of the anode feed on the water management in the cell and consequently on the current density distribution is low.

The present results show, that air dew point and air stoichiometry have a critical influence on the current density distribution in a cell of technical size. Such information is relevant for advancing the technology by improving components, such as flow fields or electrodes.

5. Conclusion

The current density distribution has been investigated in a PE-fuel cell of technical interest. For this purpose a semi-segmented plate with exactly the same properties and cell design as the one employed to build stacks of up to 125 cells, was used. Therefore, information was obtained from a cell set-up completely identical to the one having been used for construction of a fuel cell system for an automotive power train [6]. Results with respect to humidification show, that in operation modes relevant to the one of an individual cell in a stack (constant current), severe inhomogenieties in current density exist already at moderately low air stoichiometries or a moderately low air dew point. The results show, that such

measurements are a useful tool for investigation of complex parameters such as water management in PE-fuel cells.

In addition, information about the local current density can serve as an important tool to validate models. In the future, modeling will strongly support the development of improved components, such as electrodes or flow fields, when the calculated results, through extensive validation in setups of technical relevance, will be accurate and reliable.

Acknowledgment

Development of multi-channel current measurement instrumentation by R. Eckl, technical support from C. Marmy and financial support from the Swiss Federal Office of Energy (BFE) for ABG are gratefully acknowledged. All work was carried out at PSI during a leave of RPN from IST, which was financially supported by the Portuguese Foundation for Science and Technology grant no. 15805/98.

References

[1] S.J.C. Cleghorn, C.R. Derouin, M.S. Wilson, S. Gottesfeld, J. Appl. Electrochem. 28 (1998) 663.
[2] C. Wieser, A. Helmbold, E. Gülzow, J. Appl. Electrochem. 30 (2000) 803.
[3] J. Stumper, S.A. Campbell, D.P. Wilkinson, M.C. Johnson, M. Davis, Electrochim. Acta 43 (1998) 3773.
[4] M. Ruge, F.N. Büchi, in: E.W. Brooman, C.M. Doyle, C. Cominellis, J. Winnick (Eds.), Proceedings of the Symposium on Energy and Electrochemical Processes for a Cleaner Environment, 200th Meeting of the Electrochemical Society San Francisco, September 3–7, 2001, PV 2001-23, 2001, pp. 165–173.
[5] M. Ruge, F.N. Büchi, in: M. Nurdin (Ed.), Proceedings of the First European PEFC Forum, Lucerne, Switzerland, July 2–6, 2001, 2001, pp. 299–308.
[6] P. Dietrich, F.N. Büchi, A. Tsukada, M. Bärtschi, R. Kötz, G.G. Scherer, P. Rodatz, O. Garcia, M. Ruge, M. Wollenberg, P. Lück, A. Wiartalla, C. Schönfelder, A. Schneuwly, P. Barrade, Proceedings of the SATG Conference, September 13, 2002, Wil, Switzerland, ISBN 3-906483-03-7.