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ScienceDirect
Journal of Power Sources 176 (2008) 477–483
JOURNAL OF POWER SOURCES
www.elsevier.com/locate/jpowsour

Measurement of the current distribution in a direct methanol fuel cell—Confirmation of parallel galvanic and electrolytic operation within one cell

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Available online 28 August 2007

Abstract

Current production in fuel cells is typically unequally distributed along the cell surface due to inhomogeneous concentration of reactants and temperature. The inhomogeneities in fuel cells can result in reduced output power and accelerated ageing. To quantify the inhomogeneities a measurement system has been developed which allows measuring the local distribution of current and temperature in hydrogen and direct methanol fuel cells. With this system we are able to directly observe the coexistence of galvanic and electrolytic domains in a single channel direct methanol fuel cell (the electrolytic domain is the domain where electrolysis occurs in contrast to the galvanic domain where the fuel cell process takes place). The measurement device also allows for the measurement locally resolved impedance spectra.

Keywords: DMFC; Current distribution; Measurement hardware; Galvanic and electrolytic mode

1. Introduction

1.1. Non-uniform current distribution in fuel cells

Membrane electrolyte fuel cells suffer among other problems from a non-uniform current distribution within a single cell. Ideally the current density should be uniform across the active electrode surface. For different reasons, this is often not the case:

- Non-uniform loading of catalyst layers with catalyst particles.
- Non-uniform feed supply over the cell surface.
- Partial flooding of the membrane-electrode assembly (MEA) with product water.
- Blocking of MEA surface by product gases (in DMFC).
- Non-uniform temperature.

As a consequence, the output power is less than expected and due to local over-heating or high local current densities, accelerated ageing can take place. Furthermore, simultaneously occurring fuel cell regime and electrolysis regime within a single cell has been reported and experimentally confirmed [1,2].

Different constructional and operational measures can be taken to optimise the uniformity of the current distribution. These measures are mainly optimisation of air and fuel flow regimes through flow fields. But scientist and engineers need detailed data on the uniformity – or better to say non-uniformity – of the current distribution to improve the design and the operation strategies of the fuel cells.

In this work, we report the development of a measurement technique which allows measuring the current distribution within a fuel cell without disturbing the current distribution by the measurement. Beside the description of the measurement system, results on the current distribution in direct methanol fuel cells are presented.

1.2. State-of-the-art of measurement technology for current distribution

Several techniques for measuring current distributions are reported in the literature. Cleghorn et al. [3] developed one of

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doi:10.1016/j.jpowsour.2007.08.056

the first techniques of that type. They used a segmented printed circuit board (PCB), which replaced the flow field on the cathode side of a single channel PEM fuel cell. To ensure an equal potential along the channel, they used a separate electronic load with voltage sense directly on the PCB flow field for every segment.

Brett et al. [4] applied a similar design without the voltage sense to measure the local current in a single-channel hydrogen cell and they developed it further to measure the local impedance in the cell [5]. This system generally applies equal DC currents to each segment while measuring the impedance. Although this method may be good for characterising the local properties of the MEA itself, it does not take into account the local conditions like fuel concentrations and temperature. It cannot be used to measure the current distribution, as the segment current is predetermined by the measuring system.

Schneider et al. [6] discuss several methods for measuring the current distribution in fuel cells. They have found that a non-reactive measurement technique is needed. The method itself must not cause a voltage drop that influences the current. They finally used a magnetic current transducer. However, the drawback of this technology is that it is temperature sensitive and not suited for frequencies above about 1 kHz. Additional errors might result from the resistance of the wire between the segment and the transducer.

Wieser et al. [7] use an array of hall sensors and ferrites inside the fuel cell assembly to measure the current distribution. This system is only suitable for fuel cells with large active areas because of the size of the hall sensors and the ferrites. The system can also not be used to perform impedance spectroscopy measurements with high frequencies, as the hall sensors are read out sequentially within a time of at least 1 s. Furthermore the ferrites might introduce an inductance into the system, which would influence the impedance of the fuel cell significantly at high frequencies.

The company S++ [8] sells two systems for measuring the current distribution. The system "current scan lin" [9] can only measure the current in one element at a time, simultaneous measurements of several segments are not possible. In contrast the technique "current scan high res" [10] uses individual operational amplifiers to create an equal potential at each segment. The output voltage of each amplifier is a measure of the current flowing through the segment. This method would allow for the simultaneous measurement of the current in all segments. However, probably because of the high resolution (32×36 segments), the signals are multiplexed. Furthermore, the system is limited to a current of 20 mA per segment. Higher currents are available on request, but at high currents there is a non-negligible influence of the counter voltage in the wires, which connect the segments to the measurement system. This influence is not considered by this method.

Magnetotomography [11] is a different method for measuring the current distribution in a fuel cell. Here, the magnetic field around the fuel cell is used to calculate the current distribution in the cell. Although the method is completely non-invasive and can achieve a good spatial resolution, its time resolution is in the order of minutes, and therefore, not suited for impedance spectroscopy measurements.

There are several more publications on the measurement of current distribution in fuel cells [3,12–17]. However, all proposed solutions are not providing a measurement system which is not affecting the current distribution (see also Section 2.1) and being suited for impedance measurements at a time.

2. System for non-reactive current distribution measurement

In order to measure the local properties in the active area of a fuel cell, the area is divided into segments. As the fuel cell system is normally assembled between two highly conductive end plates, they can be considered as equipotential areas, and therefore, the potential difference between the endplates is the same for each point in the cell. Thus, from an electrical point of view, the only difference between the segments is the amplitude of the current and the effective resistance between the end plates, which includes the resistance due to the electrochemical processes in the cell. The measurement of the segment current must not impact this current, and therefore, the measurement should not change the segment potential.

The measuring system should thus compensate for voltage drops in the measurement device, e.g., a shunt resistor, and allow the simultaneous measurement of all segments. Additionally, the system should allow impedance measurements with frequencies up to 10 kHz. The system must operate close to a heated fuel cell; thus the temperature dependency should be as low as possible. For easy integration into the fuel cell without disturbing the flow of reactants, the measurement system should be inserted between the end plate and the flow field. For this purpose a printed circuit board (PCB) is used in design described in this paper.

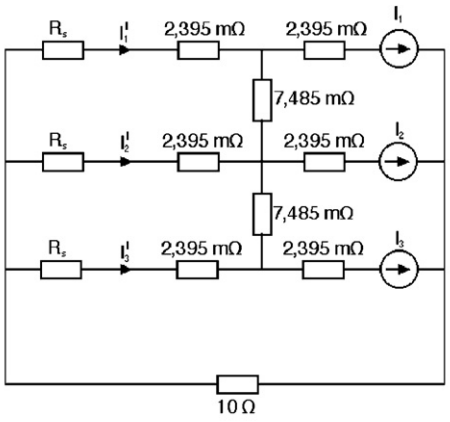
The simultaneous measurement in many (virtually all) segments provides a "snapshot" of the state of the cell. The cell (especially DMFC) is subjected to various instabilities and ageing; the snapshot gives a real picture of local parameters distribution at given operation conditions and in any point of time. Any sequential method would require statistical averaging, which would make the measurements much more time-consuming and would "mask" the physics of instability and ageing.

2.1. New current measurement system with compensated sensor resistor

Based on the above listed requirements a new measurement technique has been developed. It utilises a PCB with gold plated contacts to contact the segments of a segmented flow field.

One option is to measure the current by measuring the voltage drop over a resistor (Fig. 2). In this case the resistance has to be very small (smaller than the resistance of the segment, which is in the range of 10 mΩ for a typical laboratory cell), because the voltage drop would otherwise equal to the current distribution.

Fig. 1 shows the impact of the resistance of the shunt for current measurement in a segmented fuel cell. The simplified equivalent circuit diagram shows current sources (I'_1, I'_2, I'_3) which represent the current generation by the fuel cell process. Longitudinal and lateral resistors represent the resistance in the



R_s	I_1	I_2	I_3	I_1'	I_2'	I_3'
1 μΩ	25 mA	30 mA	25 mA	25,82 mA	28,37 mA	25,82 mA
10 mΩ	25 mA	30 mA	25 mA	26,39 mA	27,23 mA	26,39 mA
50 mΩ	25 mA	30 mA	25 mA	26,59 mA	26,82 mA	26,59 mA

Fig. 1. Simplified equivalent circuit diagram of a fuel cell and impact on current distribution depending for measurement concepts with shunts.

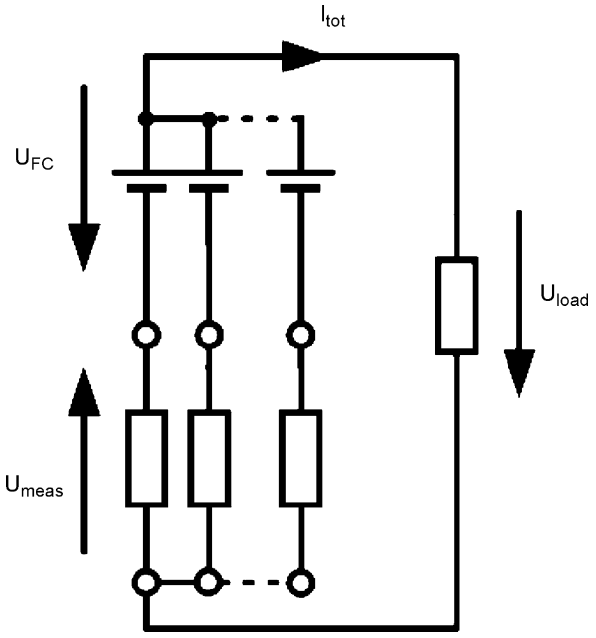


Fig. 2. Simple current distribution measurement technique without compensation.

different layers of the fuel cells. The given values are characteristic for a DMFC fuel cell as used for the investigations described in this paper. However, the example only shows the principle problems of distributed current measurements. Finally all segments are connected in parallel at the end plates, which can be treated due to their high conductivity as equipotential surfaces. Introducing the resistors R_s allows measuring the currents in each segment. But the measured currents are I_1', I_2', I_3' instead of the generated currents I_1, I_2, I_3 which are basically of interest. Due to the lateral current flows from one segment to the other the currents have a tendency to level out towards the end plates.

The table in Fig. 1 shows for a given current distribution ($I_1 = I_3 = 25$ mA, $I_2 = 30$ mA) the measured currents via a shunt-based measurement system with different resistance values. For virtually no resistance ($R_s = 1$ μΩ), the measured currents are $I_1' = I_3' = 25.82$ mA, $I_2' = 28.37$ mA. These values show that even with a perfect measurement system the real current generation can be measured only approximately. When using 10 or 50 mΩ resistors for the current measurement, the original difference in the current generation almost vanish in the light of the measurement system. Therefore, it is most important to have resistance-free current measurement to get at least the best possible result. A perfect result can be achieved only if the measurement is done directly at the point of current generation, thus, in the catalyst layer at the membrane.

Fig. 2 shows another representation for the segmented fuel cell and the current measurement system based on resistors. As already the resistance of the tracks on the printed circuit board that contact the segments is in the range of several milliohms, this technique does not allow precise measurements. Furthermore, very low resistances values result in very small signals that would need sophisticated amplification and filtering circuits. This is hardly suitable for high frequency measurements and would be sensitive to electromagnetic interference.

Therefore, a more sophisticated design has been developed. To allow for the use of a measurement shunt with higher resistance, a compensation of the ohmic voltage drop has been developed. This is done by connecting a voltage source and a transistor in series to the shunt resistor (Fig. 3). The voltage source provides a voltage that is opposite to the voltage drop of the shunt resistor during normal fuel cell operation. The amplitude of the voltage source must be large enough to level out

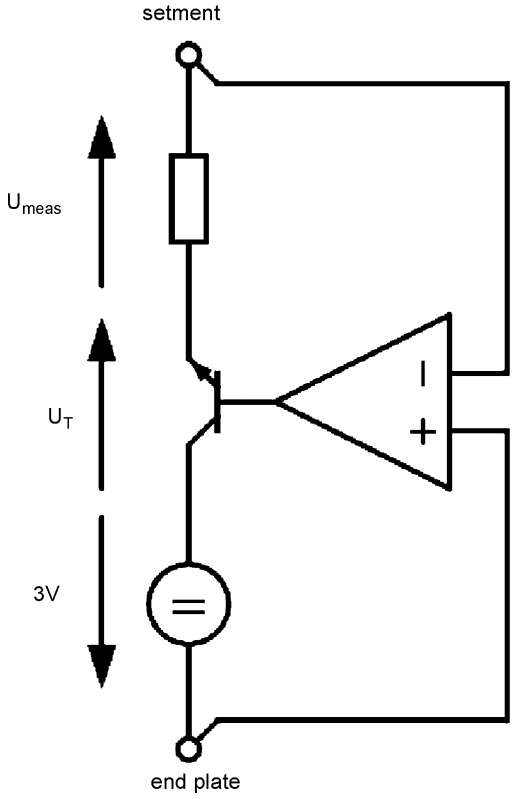


Fig. 3. Principle of the compensated sensor resistor.

the maximal voltage drop over the shunt resistor (here 1 V) and the minimal voltage drop over the transistor (about 0.7 V). It must not be too large, because the power delivered by the voltage source is dissipated in the transistor and the shunt resistor. Three volts are chosen as an appropriate value for voltage source. An operational amplifier then detects the voltage over the series connection of the three elements and regulates the transistor in a way that it levels out this voltage.

Two separate tracks connect each segment with the measurement circuit; one conducts the current while the other one provides a direct voltage sensing connection between the segment and the operational amplifier. As no current flows through the voltage sensing connection, the effect of track resistance is cancelled. According to the four-point probe method this assures that the voltage drop over the current connection is compensated. This is necessary as the resistance of this strip line on the printed circuit board (PCB) is already 10–20 mΩ, without the resistance of the connectors and the wires to the PCB with the active electronics. This method assures that the voltage between the segment and the end plate is always exactly zero, and therefore, any influence on the current distribution in the fuel cell is avoided.

Every segment is equipped with the described electrical circuit. The voltage sources can be combined into one voltage source, as the counter voltage may be the same for all segments.

When selecting a shunt resistor, one has to consider that the generated voltage must be high enough for the data acquisition system but the power losses must be limited as well. The voltage drop on the shunt resistor is amplified with a differential amplifier and referenced to the potential of the end plate. This decoupled signal can directly be recorded with a data logger or fed to an impedance spectroscope.

Resistors in a small surface mounted device (SMD) package with a temperature coefficient of less than 100 ppm K^{-1} and high precision operational amplifiers assure a good long-term stability of the system.

So far two measurement systems have been built which utilise the method described above, one with 1 Ω resistors and the other one with 10 Ω resistors, which can be used with segment currents of up to 1 A or 100 mA, respectively. The systems also allow negative currents of up to 20 mA per segment because the operational amplifier can draw small currents through a base-emitter resistor. It is also possible to build a system for symmetrical currents at full range, but this has not been realised yet. This system could be used for reversible fuel cells. Measuring higher currents in larger fuel cells is possible with the described technique as well.

Spatially resolved impedance spectra can be recorded by imposing an AC current to the fuel cell and measuring the distribution of this signal ($I_{i,\,AC}$) with the above described system. If the voltage response of the fuel cell ($U_{cell,\,AC}$) is measured at the same time, the impedance of each segment (Z_i) can be calculated using the following formula $Z_i = U_{cell,\,AC}/I_{i,\,AC}$.

2.2. Measuring the temperature distribution

Fuel cell processes are highly temperature dependent. In order to recognise temperature effects our measurement system was equipped with temperature sensors on each segment. We used digital thermometers from Maxim (Type 18B20), which have the advantage that they can share one bus that uses only one data wire. Therefore, including power supply, the whole temperature measurement system uses only three wires. Using a thermally conductive glue, each sensor is glued directly to a gold plated pin-hole passage through the printed circuit board ("via") in the middle of the segment (Fig. 4). This assures a good thermal contact to the segments of the flow field.

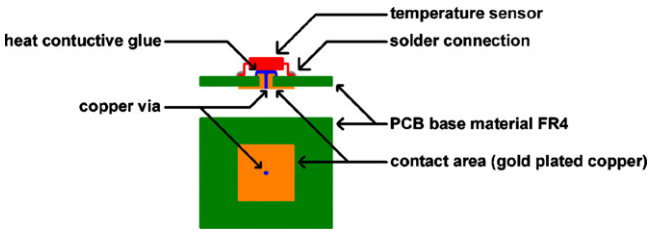


Fig. 4. Temperature sensor arrangement on the PCB [18].

3. Experimental results

The measurement system was successfully used in measurements on direct methanol fuel cells (DMFC) in collaboration with Forschungszentrum Jülich ([2,19]). The first tests of the system showed that it meets all requirements. The voltage drop over the shunt resistor is properly compensated. The system could also be used for impedance measurements with frequencies up to 10 kHz. The temperature measurement system also worked as expected.

The measurement system consists of three printed circuit boards (PCB). One is inserted between the segmented flow field and the end plate. It has gold plated contact areas for each segment on the side of the flow field. As described above, every segment is connected to the electronic circuit by two separate conductors. The other two PCBs are directly connected to the one in the fuel cell. They serve for electrical connection and hold the active electronics for the compensated shunt resistors.

3.1. Results obtained with a single channel direct methanol fuel cell

A single channel direct methanol fuel cell (DMFC, Fig. 5) was equipped with the new measurement system. The advantage of this cell is that flows in the channels are one-dimensional; this helps to understand the observed effects and simplifies their modelling. The cell consists of a 104 mm × 3 mm MEA based on a Nafion 115 membrane, which is located between two segmented flow fields and two end plates made from stainless steel (Fig. 6). The segmented flow fields are made from polysulfone with 20 embedded graphite segments of a size of 5 mm × 15 mm each (Figs. 5 and 6).

The system was used to measure the current distribution along the channel. In all experiments the fuel cell was operated at 80 °C with varied current, methanol and air flows. The temperature was controlled by a heating system. The air on the cathode side was not humidified.

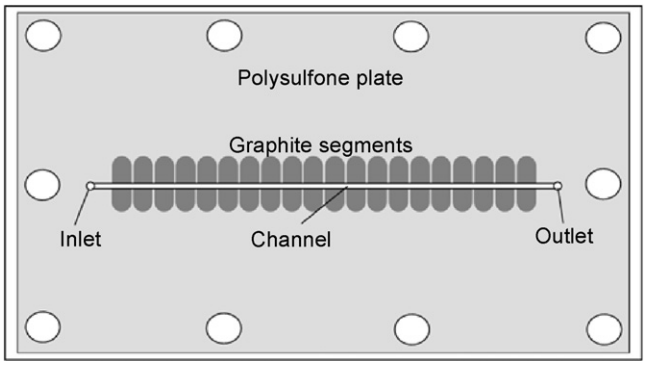


Fig. 5. Schematic of the segmented flow field with graphite segments.

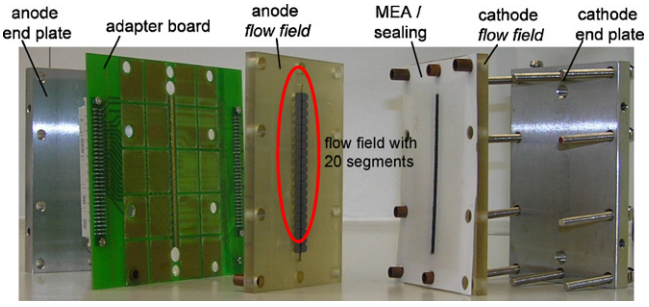


Fig. 6. Fuel cell assembly for an experimental single channel direct methanol fuel cell with measurement system adapter board and segmented anode flow field (design, construction and picture by Forschungszentrum Jülich).

The first result of this measurement was that under normal operating conditions the current is not evenly distributed over the segments (Fig. 7). The figure shows the current distribution for three different load currents. The fuel and the air inlet are at segment no. 1. The local current decreases along the cell by more than a factor of 2. Current growth close to the inlet (Fig. 7) is due to higher temperature there. The decreasing current distribution along the channel is due to a decreasing oxygen and methanol concentration in the channel. It was also observed, that the current distribution changes over time. Segments 10 and 11 seem to show a systematic error. This was found to be due to contact problems between the segmented PCB and the segmented flow field. The problem could be resolved in later measurements.

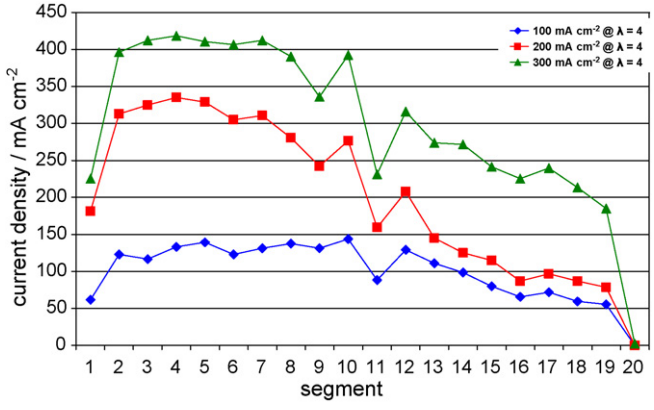


Fig. 7. Current distribution at standard operating conditions ($T = 80\,^{\circ}C$, $\lambda_{MeOH} = \lambda_{air} = 4$, $j = 100$, 200 or 300 mA cm^{-2}).

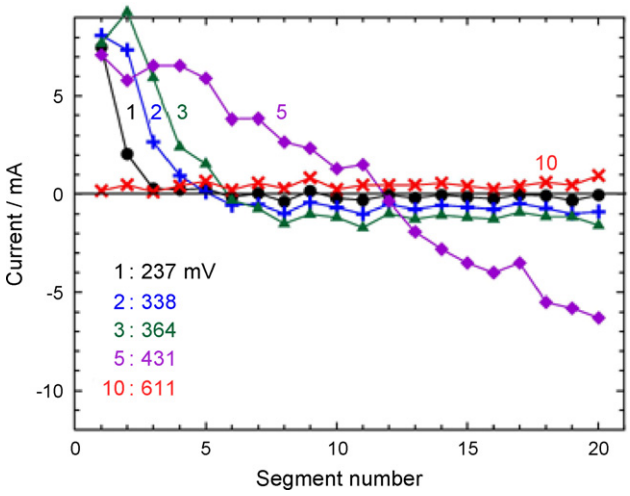


Fig. 8. Current distribution at different air flow rates (ml min^{-1}), constant current in the load of 8.5 mA and constant methanol flow of 0.1 ml min^{-1} (1 M methanol solution). Positive current indicates a fuel cell process (galvanic mode), negative currents an electrolysis process (hydrogen generation). Numbers in the lower left corner are the respective cell voltages.

The new current distribution measurement system was then used to verify an effect reported in Refs. [2,19–21]. Under certain circumstances part of the segments work in electrolysis regime and consume current instead of producing it. The first experimental evidences of this regime of DMFC operation was obtained from the measurement of hydrogen evolution, which should not be present in DMFC under normal operating conditions [21,22].

With the new measurement system, the features of this regime could be observed directly. Fig. 8 shows the measured current distribution at constant cell current of 8.5 mA, constant methanol flow rate of 0.1 ml min^{-1} (1 M methanol solution, $\lambda_{MeOH} = 114$) and different air flow rates, which are indicated in ml min^{-1} with the numbers next to the graphs. Note that the cell current of 8.5 mA is very low, which means that the cell is operated near open circuit conditions, where most oxygen is consumed for the direct oxidation of permeated methanol on the cathode, leading to very high lambda-values. At a high air flow rate of 10 ml min^{-1} ($\lambda_{air} = 65$) the current is nearly evenly distributed over all segments (Fig. 8). But at a lower air flow rate (5 ml min^{-1}, $\lambda_{air} = 33$) the cells near the air outlet (outlet at segment no. 20) start consuming current (Fig. 8). In most segments this leads to a very high local current density compared to the case of an equally distributed current. For an air flow rate of 5 ml min^{-1}, at the first segment this factor is more than 16. At lower air flow rates more segments turn into electrolysis mode. Note that at an air flow rate of 5 ml min^{-1} the current generated in the "galvanic" segments is 47.4 mA, while the current used in the electrolytic segments is 33.7 mA. The latter value exceeds the total cell current by a factor of 4–5.

The data presented in Fig. 7 can also be used to verify the function of the measurement system. Although the electronics were built for currents of up to 1 A per segment, the sum of the measured segment currents (8.3, 8.6, 9.9, 13.7 and 8.9 mA) are close to the total cell current and the resulting error of the current

measurement is less than 6.5%. The error has been calculated according to the formula $\varepsilon = \sum_{i=1}^{20}(I_i - (I_{cell}/20))/\sum_{i=1}^{20}|I_i|$ where I_i are the segment currents and I_{cell} is the cell current. The number of segments is 20.

Although this could be improved with a design of the electronics that better suits such low currents, this means that the system already works accurately at low currents.

We assume that the open circuit voltage in the "electrolytic" segments is close to zero. Since the end plates are equipotential, this causes a reversal of currents. We are currently developing a device to measure the half-cell potentials in each segment.

3.2. Local current distribution in a square-shaped direct methanol fuel cell with a grid-structured flow field

The measurement system was also used with a square-shaped direct methanol fuel cell with a grid-structured segmented flow field and 25 segments. The flow field is made of a graphite plate of 5 mm thickness, with a grid structure of 1 mm deep and 1 mm wide channels. This allows a two-dimensional gas or liquid flow in the flow field.

Fig. 9 shows the distribution of the local current density. The air inlet for this cell is located at the top left corner of the flow field slightly above the active area of the second segment from the left and the methanol inlet is located at the bottom right corner slightly under the active area of the second segment from the right. The current can be as low as 30% or as high as 144% of the expected average value. The reasons for the non-uniformity of local current density are currently under investigation. The low current is most probably caused by local accumulation of CO_2 bubbles or by local MEA flooding with product water. The current distribution is not stable with time even though the operational parameters (temperature and flow rates) are not changing.

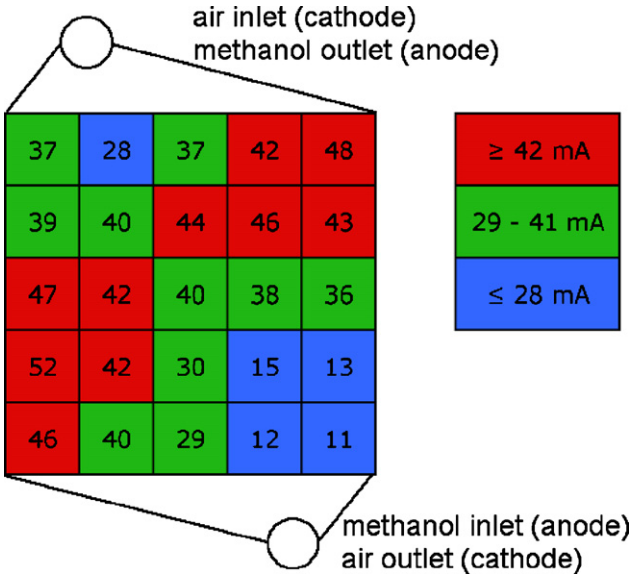


Fig. 9. Current distribution in the square shaped DMFC. Numbers indicate local current (mA). Average segment current is 36 mA. Conditions: 80 °C, air flow rate 64 ml min^{-1} ($\lambda_{air} = 4.3$), methanol (1 M) flow rate 0.4 ml min^{-1} ($\lambda_{MeOH} = 4.3$), cell current 900 mA.

4. Summary

Non-uniform current distribution in polymer electrolyte fuel cells reduces output power and accelerates ageing. Therefore, detailed information on the current distribution is necessary to optimise the fuel cell design and the operating strategies.

A measurement system has been developed which allows a precise measurement of the current distribution in linear and square-shaped fuel cells. The feature of the measurement technique is that it is non-reactive with regard to the current distribution. This has been achieved by counterbalancing the voltage drop along the measurement shunt with a constant voltage source and an operational amplifier.

Using the new measurement technique and a specially designed single channel direct methanol fuel cell, the bifunctional regime of DMFC operation was directly observed for the first time. In this regime, part of the cell works as a normal fuel cell and the remaining part operates as electrolyser. The onset of this regime depends on the air flow rate. At low flow rates, the electrolysis process is very severe.

Beside the single channel, a conventional square shaped cell was segmented into 25 elements and the current distribution was measured as well. Critical areas in the cell with reduced current production can be identified easily.

The new measurement system allows precise measurements of current distributions in hydrogen and direct methanol fuel cells. Based on this technique, the important results have been obtained already during the first measurements. This technology could provide a significant support for further developments and improvements of fuel cells.

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