



Short communication

Influences of current collector foils with different opening ratios in passive polymer electrolyte membrane fuel cells

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ABSTRACT

Even if many fuel cell applications are ready to start into the market, more research needs to be done to improve the currently achieved power density further. In the power range of about 10–20 W micro-PEM fuel cells have a high improvement potential concerning the current collector design and the design of the passive air supply. These two points have a high impact on the water management of a PEM fuel cell and allow a significant decrease of the fuel cell system in size and weight. The current work shows calculations for the fuel cell impedance based on a mathematical resistance model which was already presented for similarly constructed direct methanol fuel cells (DMFCs) [4]. Selected publications on water uptake and membrane humidification for the used Gore MEAs [6,7] are taken into account. The model is evaluated with realized versions of cathode side current collector designs, which influence the maximum power density and the self-heating of the fuel cell stack. Several measurement results are presented, which can confirm the validity of the used model. A very low opening ratio of less than 0.1 induces a very high concentration gradient of the generated water in relation to the net water outtake. From this it follows that the cell impedance is very low and the membrane has a very high ionic conductivity. Additionally it can be shown that the power density of these fuel cells is twice as high as for the cells with an opening ratio greater than 0.45.

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1. Introduction

Due to previous work at the Technische Universität Berlin [1] and taking into account other literature data and models [2,3], current collectors (Fig. 1) normally have been designed with a very high opening ratio of about 0.8 that ensure a high oxygen and water transport ability. The author's research aim was to reduce the ohmic losses of the used fuel cells by varying the opening ratios of the cathode side current collectors in a fuel cell stack with passive air supply. A mathematical resistance model was already presented for similarly constructed direct methanol fuel cells (DMFCs) [4] and is now applied to hydrogen driven PEM fuel cells as well as previously optimized structures to realize a sufficient passive air supply with also sufficient and homogeneous contact pressure application [5]. Unlike DMFCs a PEM fuel cell shows a significant dependence on the humidification of the membrane. Thus good water management is necessary. It is a challenging issue for the focused portable micro-fuel cell systems to use only the generated reaction water,

because the passive air supply does not allow gas humidification. Additionally the hydrogen is supplied with almost no water content to permit a very low amount of external peripheral components.

2. Theory

The fuel cell performance depends on different loss mechanisms, which reduce the fuel cell voltage according to Eq. (1), where U_{Rev} is the thermo neutral reversible cell voltage, η_{OC} is the open circuit over voltage, which is mainly induced by hydrogen and oxygen crossover and the relating mixed potentials. η_A is the activation over voltage, which is induced by the chemical reaction kinetics and η_{Ohm} is the ohmic over voltage, which is induced by all occurring resistances, including the membrane's ionic resistance. The values of U_{Rev} , η_{OC} and η_A are defined by electrochemistry and the MEA which is not modified. Only η_{Ohm} is investigated in this work as it is directly influenced by the current collector design. The impact of concentration losses which is also influenced by current collector design is also observed. Concentration losses are covered by U_{Rev} (by Nernst equation [9]) and η_A (by reducing the exchange current density [9]) and will be the subject of another study.

$$U_{Cell} = U_{Rev} - \eta_{OC} - \eta_A - \eta_{Ohm}; \quad \eta_{Ohm} = IR_{Cell} \quad (1)$$

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Nomenclature

κ	ionic conductivity (S m^{-1})
ρ	specific resistivity ($\Omega \text{ m}$)
c	concentration (mol m^{-3})
μ	chemical potential (J mol^{-1})
D	diffusion coefficient ($\text{m}^2 \text{ s}^{-1}$)
R	universal gas constant ($\text{Pa m}^3 \text{ mol}^{-1} \text{ K}^{-1}$)
T	temperature (K)
J_{anod}	anode side water exhaust (mol s^{-1})
J_{cathod}	cathode side water exhaust (mol s^{-1})
J_{prod}	produced water due to the electrochemical reaction (mol s^{-1})
J_{ED}	water transported by electro osmotic drag (mol s^{-1})
J_{BD}	water transported by back diffusion (mol s^{-1})

$$R_{ionic} = \frac{t}{(\kappa_{membrane} A_{geo})}; \quad t, A_{geo} \text{ membrane thickness and area} \quad (4)$$

$$\kappa_{membrane} = t_1 \phi^{t_2}; \quad t_1 = 7.475 \text{ S m}^{-1}, \quad t_2 = 1.74 \quad (5)$$

The current collector bulk resistance R_{along} and the resistance of the diffusion layer $R_{GD Liz}$ are calculated numerically in 3D using FEM tools because of the non-linear current distribution. Assuming a homogeneous current distribution (red arrows in Fig. 2) a simplification can be made for R_{along} which allows a more analytical modeling approach. This approach yields R_{along} according to Eq. (6) with the standard bulk resistance depending on the current collector's length L and cross-section (wh) extended by a factor $1/3$, which represents the 90° current flow. This can be derived analytically using an integral approach for the infinitesimal power loss IR' . The factor $1/4$ is used, because the load is electrically connected with one contact on each side of the current collector (green arrows in Fig. 2), so that the current effectively is drawn through two symmetrical parts of half the length ($=1/2$) which are electrical in parallel ($=1/2$ again).

$$R_{along} = \frac{1}{4} \frac{1}{3} \frac{\rho L}{wh} \quad (6)$$

The diffusion layer resistance $R_{GD Liz}$ can be calculated with similar assumptions (not shown here). The results are given in Fig. 3. The model uses a constant ionic resistance for a relative humidity of 0.45 (with parameters from [6]). At the moment it is not clear what humidity can be assumed for the membrane adjoining gas phase. In the literature it is often 1, also because it is assumed that the presence of liquid water is the source of membrane wetting. Nevertheless, the diagram illustrates a strong discrepancy as the measurements indicate a non-constant value of the ionic resistance for different versions of fuel cells with varied width of current collector contact bars. Fig. 3 shows the membrane's ionic

A resistance model is developed which represents all electrical and ionic resistances (Fig. 2, Eqs. (2) and (3)). The electrical resistance consists of the current collector's bulk resistance R_{along} , the contact resistance between the current collector and the diffusion layer (GDL) R_{cont} and the resistance of the diffusion layer itself $R_{GD Liz}$. The contact resistance and the bulk resistance of the catalyst layer are neglected. The resistance R_{ionic} of the membrane is approximated based on the literature [6] according to Eqs. (4) and (5).

$$R_{Cell} = R_{electric} + R_{ionic} \quad (2)$$

$$R_{electric} = R_{along} + R_{cont} + R_{GD Liz} \quad (3)$$

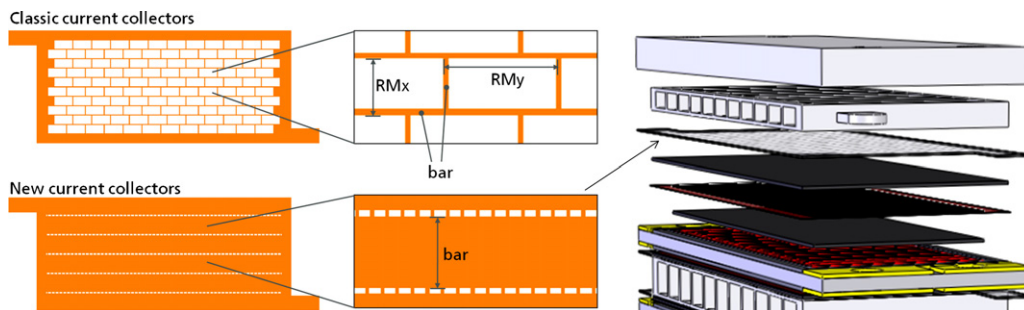


Fig. 1. Different designs for current collectors and placement in a fuel cell stack.

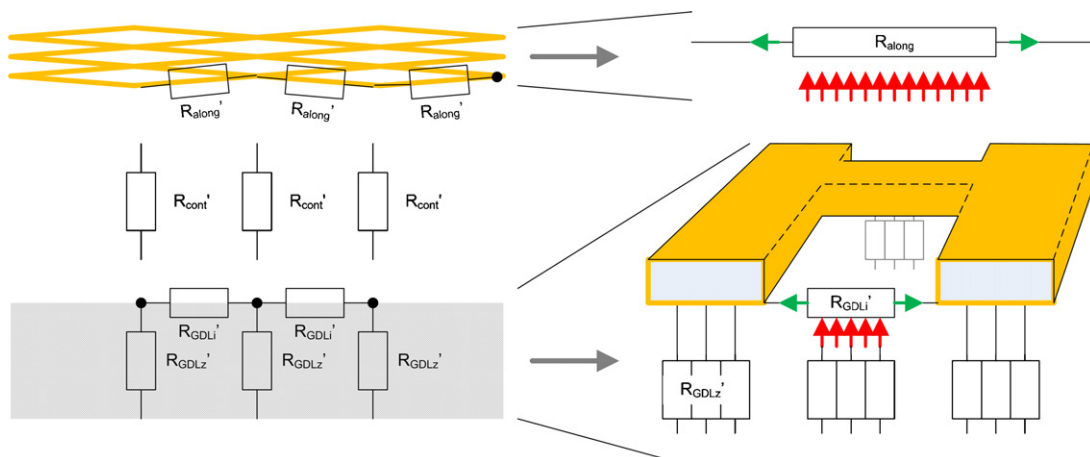


Fig. 2. Resistance model with current collector and diffusion layer.

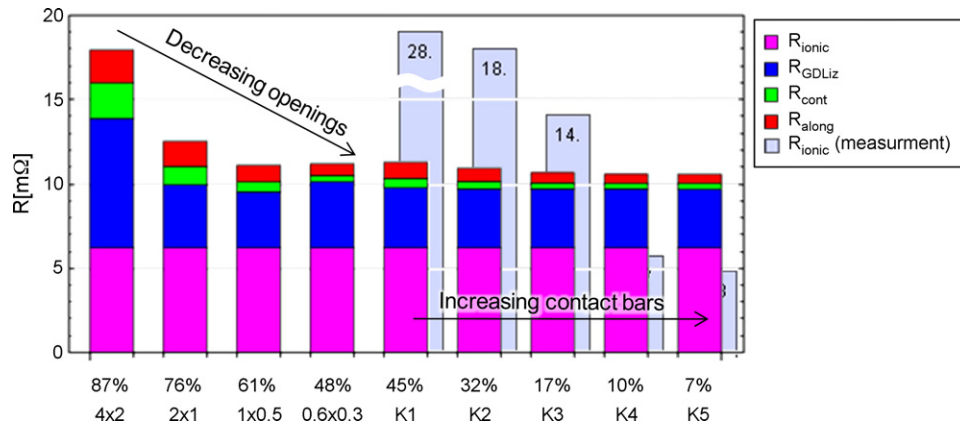


Fig. 3. Resistance model results for an active cell area of 20 mm × 50 mm.

resistance measured during a current load of 100 mA cm^{-2} , which demonstrates a very high dependence of the ionic resistance on the current collector's contact bars, which is discussed in the next section (Fig. 4).

2.1. Water model and membrane resistance

Under open circuit conditions the membrane conductivity depends directly on the ambient water activity φ (relative humidity). However, when the fuel cell is under current load, water is generated at the cathode according to Faraday's law, which results in several concentration gradients. This can induce higher membrane conductivity than water-saturated air would do. This behavior is observed in measurements and outlined in Fig. 5. All these effects are strongly influenced by the design of the current collector's openings. It is the objective of this work to better understand these effects with the help of a 1D simulation of the water distribution.

The current understanding is based on Fick's law of diffusion according to Eq. (7) with the net water flux J , the diffusion coefficient D , the concentration c and the diffusion length dx , which is equal to half the width of the current collector's contact bar. When the anode side gas channels are used in dead end mode with a very small amount of purging gas exhaust the net water flux can be assumed to be equal to the produced water, which is equivalent to the electrical current according to Faraday's law. Also the electro osmotic drag J_{ED} becomes equal to the back diffusion J_{BD} . This is

expressed in Eq. (8) as pointed out in Fig. 5.

$$J = -D \frac{dc}{dx} \quad \text{or} \quad J = -D \frac{c}{RT} \frac{d\mu}{dx} \quad (7)$$

$$J_{Anod} \rightarrow 0 \Rightarrow J_{Cath} = J_{Prod}, \quad J_{ED} = J_{BD} \quad (8)$$

The diffusion coefficient is a material parameter not exactly known at the moment. It strongly depends on the porosity of the gas diffusion layer and the electrode catalyst layer as well as on the water transport capability of the electrolyte phase. It is also part of current discussions whether to consider the concentration gradient dc (Eq. (7), left) or the gradient of the chemical potential $d\mu$ (Eq. (7), right) to calculate the water flux. As it is well known, that the chemical potential of water (liquid or gaseous phase) is equal in two domains, when the system is in thermodynamic equilibrium, the chemical potential approach allows to measure the membrane impedance in different ambient humidity states and adopt the results for steady load states afterwards. This is currently under investigation and topic of another study. So far the influence of the discussed behavior on the fuel cell performance is presented in Section 3 as empirical state of the art.

3. Experimental

Several fuel cell stacks were built with different current collector designs. The stacks are based on so-called bi-cells, where two fuel cells are assembled in opposite directions based on the same substrate, which provides the anodic flow field and current collector. The active area of each cell is $20 \times 50 \text{ mm}$ and 6 cells are placed in one stack. In principle the model can be applied to fuel cells of any size, with the condition that the cell length and width are greater than (at least two times) the width of the current collectors contact bars. The second condition is that the number of cells in the open cathode technology stack should be greater than in order to have a sufficiently homogenous temperature profile over the stack and almost all have similar temperatures.

The cathode side current collectors are etched aluminum foils plated with $3 \mu\text{m}$ nickel, $2 \mu\text{m}$ silver and $0.4 \mu\text{m}$ gold. Different versions were used having opening ratios from 0.03 to 0.45. The openings are having a constant size of $0.3 \times 0.6 \text{ mm}$, the contact bars are varied as noted in Table 1.

The preliminary experimental membrane electrode assembly Gore MX815 with $0.1 \text{ mg Pt cm}^{-2}$ catalyst coating on the anode side and $0.4 \text{ mg Pt cm}^{-2}$ on the cathode side respectively is used for all the built fuel cells. For the diffusion layer SGL Sigracet 35ac is used on the anode side and Sigracet 35cc on the cathode side. In the passive cell design an additional component with $4 \times 4 \text{ mm}$ channels is used to supply ambient air via natural convection as

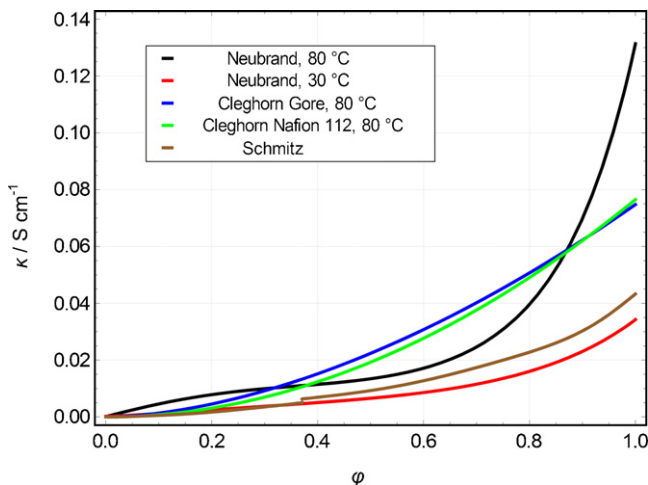


Fig. 4. Membrane conductivity taken from [6–8].

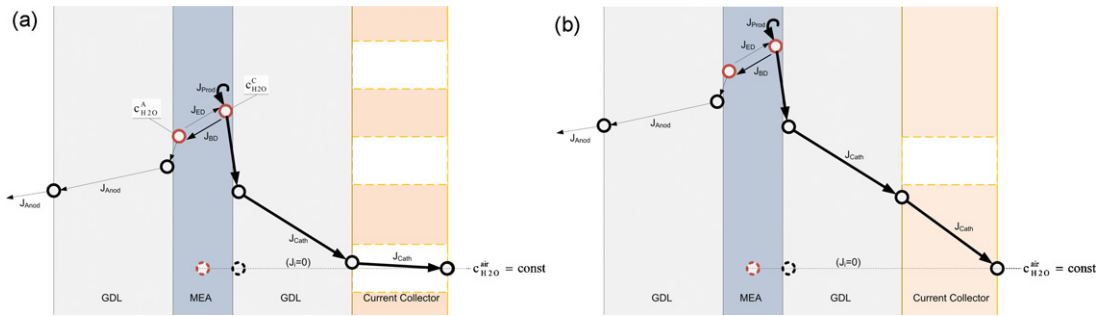


Fig. 5. Water concentration gradients (in principle, not to scale), (a) high current collector's opening ratio, (b) small opening ratio. The dashed line shows the membrane wetting due to the ambient water activity.

Table 1
Relevant dimensions of the cathode side current collectors.

	Design							
	K1	K2	K3	K4	K5	K6	K7	K8
Contact bar [mm]	0.2	0.4	1.0	1.9	3.0	3.8	4.7	6.3
Opening ratio [%]	45	32	17	10	7	5.5	4.5	3.4

well as forced air using a standard 45 mm fan to the fuel cell cathode. The stacks were compressed by the endplate mountings with a pressure of about 0.7 MPa, which is sufficient to achieve a good electrical contact. If the compressive changes to GDL diffusivity are big enough to influence the optimum current collector opening ratio has not been investigated yet. However, as all of the studied cells were compressed in the same way, this influence is not affecting the impact of the current collector design. The construction principle is illustrated in Fig. 1.

The fuel cell stacks are investigated under constant ambient conditions at steady state loads for different current values. Additionally several polarization curves were measured.

At a typical working point of fuel cells at 650 mV (Fig. 6, grey dashed line) a power density of 200 mW cm⁻² can be achieved for the fuel cell using the cathode side current collector K5 with a low opening ratio. Compared to the current collector K1 (Fig. 6, marked with circles) this is twice as high. Concentration losses are becoming visible for the fuel cells with the largest distance between air openings (K6–K8). The concentration overpotential probably mostly depends on the limited diffusion rate of oxygen inside the gas diffusion layer with the diffusion length modified by the width of the current collectors contact bars.

Fig. 7 shows plots of stationary results as function of the width of the contact bars of the cathode side current collectors. It can be

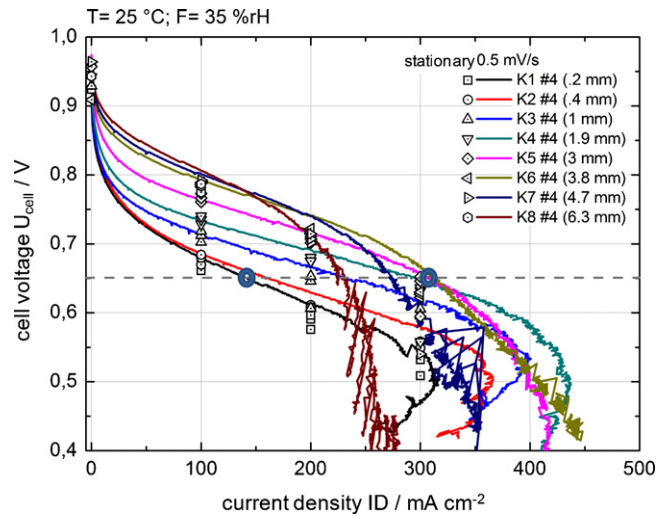


Fig. 6. Polarization curves.

seen that an increasing width of the current collector's contact bars results in a significant reduction of the cell impedance. Also a corresponding increase of the power density can be observed with a maximum for contact bars with a width of 3–5 mm, depending on the load current density. Impedance measurements of the cells with the different current collector designs (varied R_{along}) are showing only small differences for the cell resistances. Therefore it is valid to assume that the impedance reduction occurs only under load currents when the diffusion phenomena denoted in the theory chapter becomes a relevant effect.

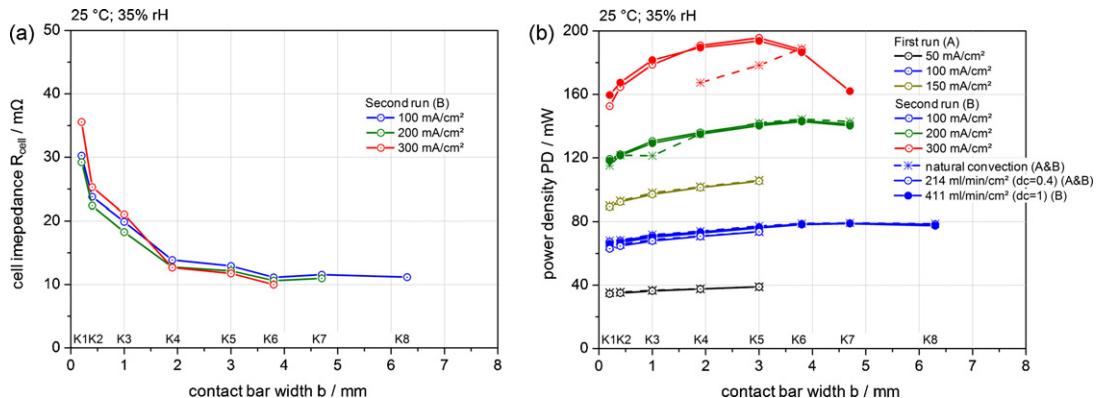


Fig. 7. Results: impedance (a) and power density (b) of a centre cell.

4. Conclusion

The current work shows a significant influence of the design of cathode side current collectors on the fuel cell impedance and the resulting power density of semi-passive PEM fuel cells. A very low opening ratio of 0.07 induces a high concentration gradient for the generated water in relation to the net water outtake. This results in a high water concentration in the membrane and consequently a high ionic conductivity. It can be shown that the power density of a fuel cell with 3 mm wide contact bars for the cathode side current collector (i.e. 0.07 opening ratio) is twice as high as for cells with an opening ratio of 0.45. Furthermore a positive feedback can be observed. The reduced self-heating keeps the air near the membrane at moderate temperatures. This in turn leads to a higher water activity in the air close to the cathode keeping even more water within the cell and maintaining a low membrane impedance.

Impedance calculations were performed based on available publications on water uptake and membrane humidification of the used commercial MEAs [6,7] in addition to the electrical resistance model. The current results indicate the need for a model that implements the interaction of the water and oxygen concentration as function of the current density and the width of the current collector's contact bars.

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