

Fluid Flow and Current Density Distribution in Large-Area HT PEMFCs

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Abstract: High temperature polymer electrolyte membrane fuel cells (HT PEMFCs) are very promising technologies when used in combined cooling and heating power (CCHP) systems. They are operated at 160°C, offering the possibility of high tolerance to fuel impurities and a possibility to use the heat generated for cooling and heating purposes, leading to higher total system efficiency. Employing a 24-channel gas supply (300 cm² of active area), fuel and oxidant gas velocities in the flow field and the porous media and their pressure profiles were studied. The electrochemical behaviour studied using various techniques is compared to COMSOL Multiphysics® based simulations. Optimum operating conditions, based on even current distribution (across the entire active area) and lowest possible pressure drop were identified.

Keywords: Fluid Flow, HT PEMFC, Current density

1. Introduction

Fuel cells should play an important role in forthcoming power systems and power supply. In the last decades, researchers heavily focused on new fuel cell layouts, optimized designs and continuously improved the durability. Because of the high aspect ratio (different length scales from e.g. 1·10⁻⁶ m to 1·10⁻¹ m) between the different components of the fuel cell, only few quantities can directly be measured during operation. Beside the currently used in-situ measurement techniques, computational fluid dynamics (CFD) became an important tool in research and development in order to simulate the performance of a specific layout before actually manufacturing it.

The aim of the work is to analyse electrochemical performance by studying fluid flow patterns and cell performance at different load currents in a large area (300 cm² of active area) HT PEMFC. Fuel and oxidant gas velocities in the flow field and the porous media and their pressure profiles do directly affect the cell performance. The resulting

electrochemical behaviour was studied using techniques such as EIS (Electrochemical Impedance Spectroscopy) and was compared to COMSOL Multiphysics® based simulations. Moreover, the issues associated with operating and modeling large area cells are discussed. Special emphasis is put on modeling approach and resources.

2. HT PEMFC Prototype Layout

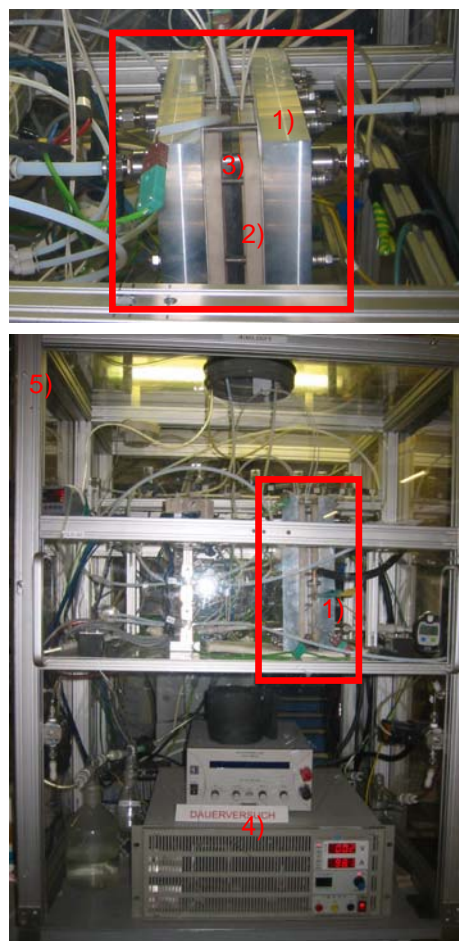


Figure 1. Tested HT PEMFC prototype and test stand. 1) Endplates, 2) High temperature resistant PEEK plates reinforced with glass fibre, 3) High temperature resistant bipolar plates, 4) Electrical loads, 5) Test stand.

The cell's active (catalyst and membrane) area was 300 cm^2 and the cell's total geometrical area was 503 cm^2 . The cell's performance was studied by operating the same in a test stand containing the layout shown in Fig.1.

Firstly, the cell was constructed with the essential parts shown in Fig.1. The endplates are made of Aluminium combined with high temperature resistant PEEK plates reinforced with glass fibre. Heater pads were inserted into the end plates of the fuel cell and their terminals were connected to the electricity supply mains in the laboratory. The bipolar plates are of graphite compound based high temperature stable material containing a 24-channel parallel serpentine type (divided into two blocks or parts of 12 channels each) flow field channels machined on them (Fig.3).

When highly doped membrane containing membrane electrode assemblies (MEAs) are employed in a HT PEMFC, typically the ohmic resistance or high frequency resistance of the cell would be low compared to a lowly doped one. However, cell kinetics are a function of catalyst loading, structure, catalyst utilization and operating temperature. In the current work, a lowly doped MEA, containing about 8 moles of phosphoric acid (PA or H_3PO_4) per mole of the polymer repeat unit and a platinum (Pt) loading of $1 \text{ mg}\cdot\text{cm}^{-2}$, each on the cathode as well as on the anode side was employed.

3. Test Stand Layout and Experimental Measurements

The test stand was constructed with mass flow controllers (MFCs) and a solenoid valve connected to it. When any MFC is not used, the connected valves were automatically closed, thus not allowing any back flow into the MFCs. Additional safety valves ensured that the inlets of the fuel cell are closed when the cell was not in operation, thus safe guarding the electrolyte from being affected by the connected pipes and the media in contact. Similarly the valves were fixed to the outlets of the fuel cell. The electronic load which has the possibility to measure voltages close to 0 V was connected to the fuel cell as depicted in the test stand layout (Fig.1).

As for the safety, various protocols were implemented during start up, shut down and on load operation regimes of the fuel cell, depending on the tests performed. For instance, while feeding the fuel and oxidant to

the fuel cell, care was taken not to exceed a differential pressure of $\sim 300 \text{ mbar}$ (across both sides of the cell). The safety protocol implemented ensured that in the event of any excess differential pressure across the fuel cell MEA, the gas supply was cut off, load was disconnected, the in-, and outlets of the fuel cell were closed and about 1 litre of N_2 gas was allowed into the anode of the fuel cell (to bring down the cell voltage level from its open circuit potential). In case of any gas leaks, the sensors installed in the test stand (very close to the fuel cell) would send an electrical signal, thus implementing a safety protocol. The outlets of the anode and cathode sides of the HT PEMFC were directed through a two stage condenser system where most of the water was collected. Water was removed from these bottles and was transferred into other clean bottles which were sufficiently rinsed with deionized water and were labeled accordingly, before they were analyzed to find out elements present in the water.

After applying a compacting force of about $1.5 \text{ N}\cdot\text{mm}^{-2}$ on the cell assembly, the cell was heated to 160°C from room temperature in 40 minutes clock time using flat heating pads inserted in the endplate on the inside (close to the current collector plate). The cell was initially operated with hydrogen (H_2) and oxygen (O_2) with gas feeds of $300 \text{ ml}\cdot\text{min}^{-1}$ and $1000 \text{ ml}\cdot\text{min}^{-1}$ on $100 \text{ mA}\cdot\text{cm}^{-2}$ of load at 160°C for about 100 hours.

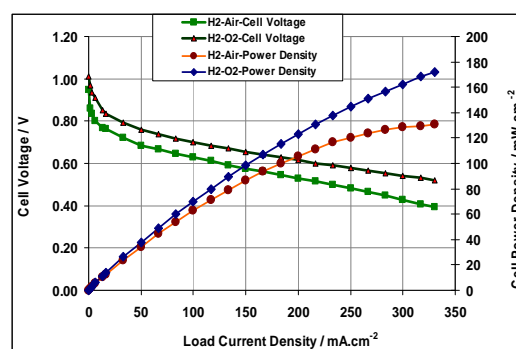


Figure 2. Recorded performance curves of the HT PEMFC prototype.

After this period, the cell performance was studied with H_2 and air feeds with stoichiometries of 1.35 and 2.5 respectively as well as with H_2 and O_2 with stoichiometries of 1.35 and 2.5 respectively, up to a load current of 99 amperes ($330 \text{ mA}\cdot\text{cm}^{-2}$) (Fig.2). The same procedure was repeated at temperatures of 160°C to 190°C and at regular intervals from 0 to $330 \text{ mA}\cdot\text{cm}^{-2}$. It must be noted that

the cell was held at different load currents for about 30 minutes clock time to enable the cell to find its equilibrium with its surroundings at that temperature, load current and gas feed rates. The cell's voltage was recorded by the computer connected to the test stand equipped with LabView® from National Instruments (NI), USA.

The cell's high frequency resistance was obtained from the high frequency intercept of the Nyquist plots obtained by the electrochemical Impedance spectroscopy (EIS) device from Zahner Messtechnik (IM6), Germany. The impedance spectra at different load currents were obtained at cell operating temperatures of 160°C to 190°C in the galvanostatic mode, by supplying scanning signals in the 100-600 mA range (RMS values). The scanning frequencies were from 1 Hz to 50 kHz.

From the spectra obtained, the high frequency x-axis intercept of the Nyquist plot was taken as the ohmic resistance of the cell. Furthermore, impedance spectra were obtained at a very low load current of 0.5 Amps (1.66 mA·cm⁻²). From the Nyquist plots drawn, the charge transfer resistance was obtained, namely R_{ct} . From this value, the exchange current densities i_0 representing cell's activation loss (of both anode and cathode side) were ascertained (see Table 1).

Table 1: Exchange current densities

T °C	R_{ct} Ω	$I_{0,c}$ A	$i_{0,c}$ mA·cm ⁻²
160	0.014634	1.275198	4.25
170	0.014525	1.314455	4.38
180	0.014395	1.356246	4.52
190	0.014099	1.415296	4.72

Beside the above measurements, the pressure drop across the anode side as well as on the cathode side were continuously recorded by the pressure sensors connected both to the in-, and outlets of the cell assembly. The recorded pressure drop values were compared to the values obtained from simulations performed with COMSOL Multiphysics®. The measured exchange current densities were used to iteratively ameliorate the model set-up.

4. Modeling and Simulation – Modeling Geometry, Subdomain and Boundary Equations

This work presents a three-dimensional, steady-state, non-isothermal model of the HT PEMFC prototype using a PA doped polybenzimidazole (PBI/PA) membrane. The model accounts for the flow fields, the gas diffusion layers (GDL) and the membrane. In this set-up, the reaction layers are treated as thin two-dimensional layers. The computational geometry, shown in Fig.3, was generated using two-dimensional drawings, extruded to the third dimension (z -coordinate).

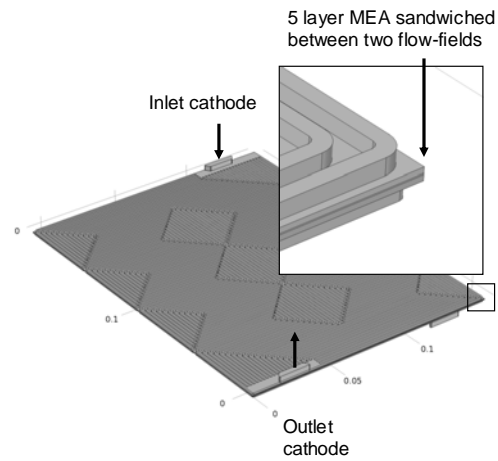


Figure 3. Three-dimensional, computational geometry in x - y - z -view (Flow field: Parallel serpentine (two times 12 channels / same on anode and cathode side)).

Overall, the governing equations are almost the same as the ones presented in [1]. The full Navier-Stokes equations were used at the anode and cathode side to model the fluid flow within the open gas channels. Within the porous media, the Brinkman equations were used. Similar to previous works, a two-phase temperature distribution was computed, separating the solid-, from the fluid-phase temperature. In total, seven application modes were used within the model set-up. The electrochemical reactions are modeled using an agglomerate approach and include the gas diffusivity and the gas solubility into PA (H₂ and air). The conductivity of the membrane is set constant.

The boundary equations were chosen according to analytical calculations and realistic operating conditions regarding load current, cell voltage, pressure, gas composition, and temperature. As mentioned, for both reaction layers, electrode-electrolyte-interface coupling boundary conditions were chosen from the dedicated 'Batteries & Fuel Cell Module' menu (including the definition of

the coupled reaction, the number of participating electrons, and the stoichiometric coefficient).

By solving a coupled system of subdomain, boundary and initial conditions along with thermodynamics, electrochemistry, and fluid-, and material properties it was possible to simulate and predict quantity distribution inside the components of the HT PEMFC prototype.

5. Use of COMSOL Multiphysics® – Mesh Details and Solver Settings

COMSOL Multiphysics® was used during the design and layout process of various cell components, namely the 24-channel gas supply (bipolar plates). The idea was to compute possible pressure drops across the flow field before even manufacturing it. After the initial experimental measurements, the model was iteratively updated using the recorded data.

All computations were performed with version 4.2.0.187 of the software using the products ‘COMSOL Multiphysics’ and the dedicated ‘Batteries & Fuel Cell Module’ (64 bit version). The used hardware was a 16-core machine with a total of 144 GB RAM.

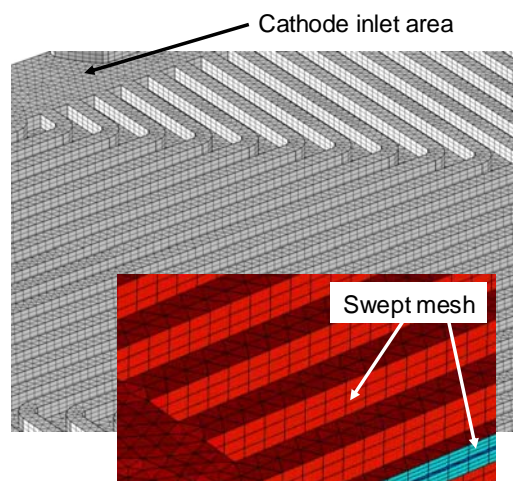


Figure 4. Three-dimensional, computational mesh in x - y - z -view showing the different types of elements.

In a next step, the complete geometry was carefully discretized while taking care of several mesh refinements required for the fluid flow modeling and simulation (for details see Fig.4 and Table 2). The entire flow fields and the GDL subdomains were meshed using a swept mesh operation, whereas the membrane subdomain was meshed with tetrahedral

elements only because both flow fields are turned 180° against each other. Before the final simulations, a mesh-independent test was performed with several dummy simulations.

Table 2: Meshing details

Type	Number of elements
Tetrahedral	758,748
Prism	2,181,778
Triangular	1,221,730
Quadrilateral	305,178
Average element quality is: 0.4343	
Total number of degrees of freedom (DOF) is: 19,692,625	

Most fluid flow computations were solved with the (parametric) direct solver MUMPS (free and porous media flow). For the transport of concentrated species and the secondary current distribution, a two-group segregated solver set-up returned best results (group 1: variables O_2 , H_2O_g , H_2 ; group 2: variables: two-phase potential). For the heat transfer the same segregated solver was used (group 1: variable T_s ; group 2: variable T_f). The model was iteratively solved while using the current solution as initial input for the next one.

6. Simulation results

The simulation results are shown for the following operating conditions: Cell voltage $U = 0.6$ V, $T_s = 160^\circ\text{C}$, $T_f = 21^\circ\text{C}$, gas flow rates according to desired load current of $I = 120$ A, stoichiometries of 1.35 (anode) and 2.5 (cathode), no backpressure. As mentioned above, the conductivity of the membrane is set constant to the value $\sigma_{MEM} = 0.08$ S·cm⁻¹. The Pt-loading of the reaction layer is 1 mg·cm⁻².

Fig.5 shows the cathode side pressure drop. A value of 52 mbar is returned. The average gas channel velocity is 6.95 m·s⁻¹. In Fig.6, the anode side pressure drop is given as line-plot for the mentioned operating conditions but for two different gas compositions. The line-plot is located exactly at the middle of the 12 channels part as indicated in Fig.5. Values of 2 and 4 mbar are returned. The average gas channel velocity is 1.75 m·s⁻¹ and 2.46 m·s⁻¹ respectively.

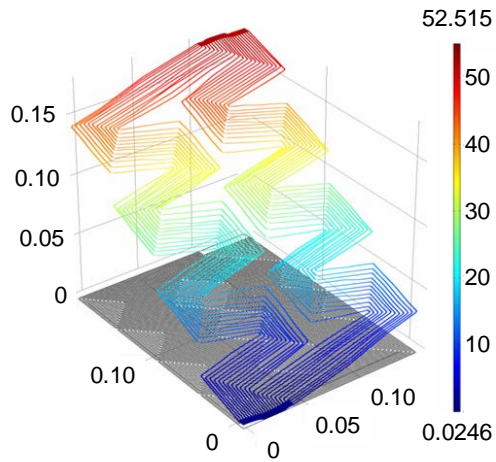


Figure 5. Cathode side pressure drop in millibar at given operating conditions (slice-plot in x - y -plane, $z = 500 \cdot 10^{-6}$ m).

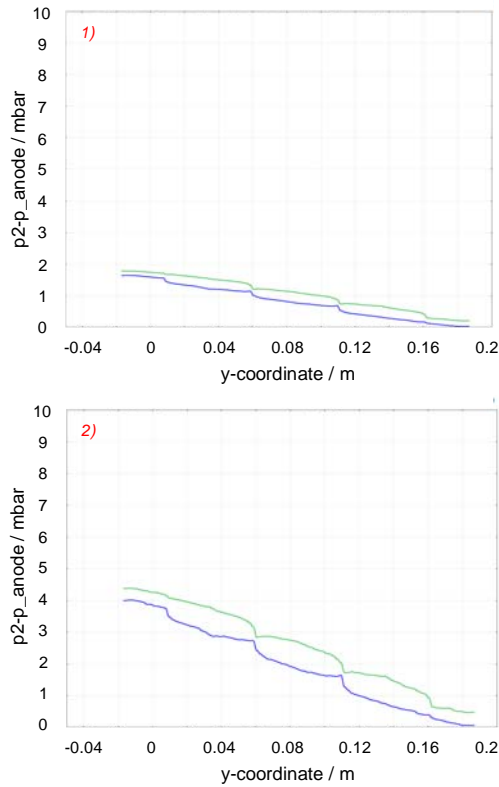


Figure 6. Anode side pressure drop (line-plot along the y -coordinate at $x = 0.04$ m and 0.096 m, $z = -600 \cdot 10^{-6}$ m). 1) at given operating conditions, 2) H_2 plus 30% fuel contaminants.

When taking a closer look at the pressure drop within the porous media, it was observed that a significant pressure difference exists in the region under each 180° bend. Consequently, a certain amount of gas bypasses the gas flow channels through the porous media. This fact may lead to higher gas

concentration of the reactants in the regions close to the reaction layer and might be a reason for higher current density values at these locations (Fig.8). A very similar behaviour was reported in [1].

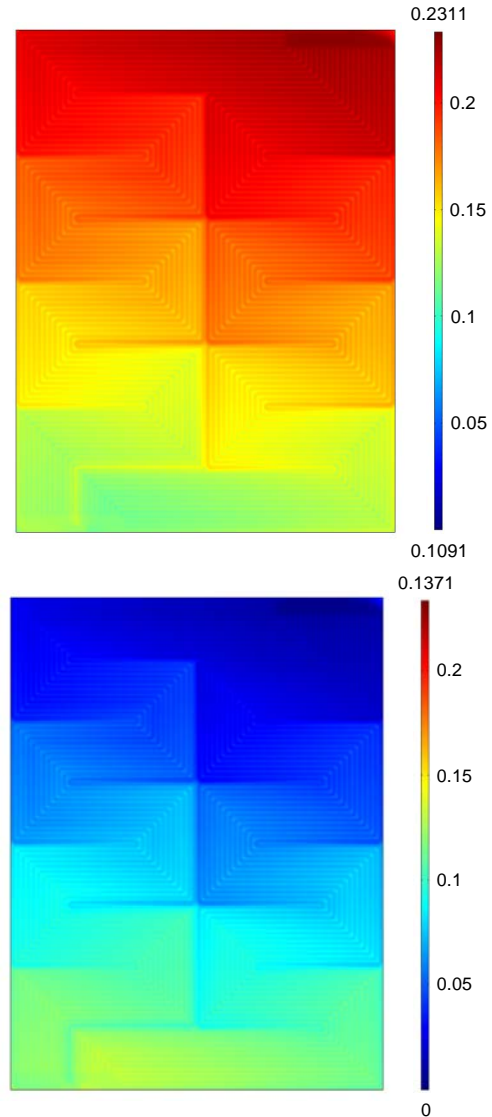


Figure 7. 1) Oxygen mass fraction at given operating conditions, 2) Water mass fraction at given operating conditions (slice plot at the GDL to reaction layer interface).

The oxygen mass fraction, the cathode side water mass fraction, and the resulting current density distribution at the GDL to reaction layer interface are shown in Fig.7 and Fig.8. The oxygen mass fraction and the current density distribution show higher values towards the air inlet and lower values towards the air outlet. The decrease in oxygen mass fraction is almost linear in along the channel

direction due to ongoing electrochemical reactions. Additionally, the current density distribution is strongly influenced by the structure of the flow field. An influence of the inlet and outlet areas is clearly seen in Fig.8. This indicates that a careful design of these locations is very important and should not be neglected. As for the cathode side water mass fraction, it increases towards the outlet. Slightly higher values are observed under the land areas. This particular distribution is of great importance when investigating H_2O/H_3PO_4 interactions in HT PEMFCs.

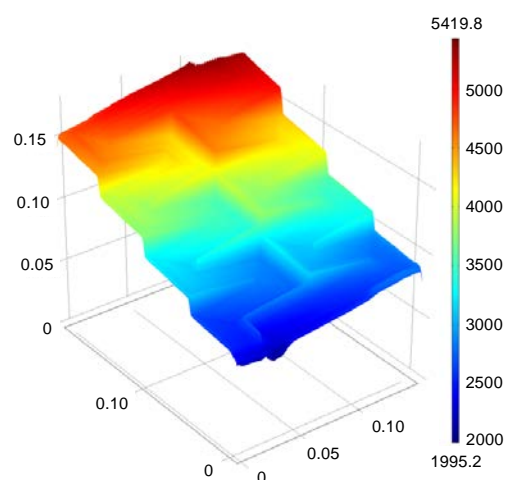


Figure 8. Current density distribution ($A.m^{-2}$) at given operating conditions (slice plot at the GDL to reaction layer interface).

The shape of the mass fractions is typical for a HT PEMFC. Overall, the simulation results agree with operating cell behaviour.

7. Conclusion

Large area HT PEMFCs pose challenges while designing and operating them. The entire active area must have uniform compression over it, to ensure durability and even current distribution. For instance, in large area cells, when the reaction of the cell components held by the end plates cause uneven thicknesses of the PA doped membrane, uneven pore size of the gas diffusion electrode (GDE), uneven contact resistance between bipolar plates and GDE as well as uneven contact resistance between current collector plate and the bipolar plates, the current distribution across the entire area will vastly vary depending on where they are located. Also, in large area cells, the cathode side outlet must ensure sufficient oxygen supply on the cathode side, where

enormous amounts of less useful gases such as nitrogen and water vapour could accumulate. The flow field plate design must address these issues, together with a low pressure drop. The mass and heat flows in cells containing large areas pose very complex challenges. Envisaged simulations demand larger computing power and memory resources to perform simulations with acceptable accuracy. The obtained results are a necessary step to compare the cell's electrochemical performance to the results obtained from experimental measurements. In the current work, there was acceptable agreement of the predicted results between and the measured values. The simulations have also yielded important and necessary quantities profiles relevant to the manifolds at the in-, and outlets of the flow field plate. Improvements on the manifolds both on the anode as well as on the cathode side are planned, also by taking other necessary parameters into account. Maintaining uniform current density distribution by ensuring adequate oxidant and fluid flow is crucial in large area cells. The simulations performed and the experimental results obtained lead us to a more improved and a stable HT PEMFC design. In the next iterations, validation will be performed and changes are implemented in the model.

8. References

1. Siegel, C., Bandlamudi, G., Heinzl, A, A Systematic Characterization of a PBI/H₃PO₄ Sol-Gel Membrane – Modeling and Simulation, *J. Power Sources*, **196**, 2735-2749 (2010)

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