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Theoretical study of the influence of material parameters on the performance of a polymer electrolyte fuel cell



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HIGHLIGHTS

G R A P H I C A L A B S T R A C T

- Material properties impacts on FC behaviour was studied by the CFD simulation.
- 25 parameters of PEM, GDL and CLs were investigated on single channel fuel cell.
- Conductivity and thickness of PEM, GDL have the most influence on FC efficiency.
- Parameters of the anode catalyst layer insignificantly affect the FC performance.

A R T I C L E I N F O

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ABSTRACT

The paper presents a systematic investigation of the influence of alterations in the values of the polymer electrolyte membrane, catalyst layers and gas diffusion layer characteristics on the performance of a PEMFC. The individual influences of 25 material properties were tested using CFD simulation on a single channel fuel cell.

The calculations of PEMFC performance were conducted by increasing and decreasing the values of each tested parameter, and comparing the results to a reference case. The dependencies of the current density on the following quantities were analysed in detail: 1) the cell potential, 2) the power density, 3) the membrane over-potential, 4) the mean water concentration in the PEM, 5) the relative humidity at the interface CCL/GDL, and 6) the total water flux through the PEM.

The results showed that the variations in the conductivities and thicknesses of the PEM and GDL, as well as variations in GDL porosity, led to significant changes in fuel cell performance. The characteristics of the anode catalyst layer had little influence on fuel cell behaviour. Increasing the thickness and exchange current density of the cathode catalyst layer increased the current densities, while the reduction of the transfer coefficient decreased fuel cell performance.

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1. Introduction

Over the past decade, Polymer Electrolyte Membrane Fuel Cells (PEMFCs) have been intensively studied due to their high efficiencies and low emission levels. Numerous parameters influence the performance of the fuel cell, whereby each parameter change affects fuel cell efficiency in a different way. To design the fuel cells,

Abbreviation: 1D, one-dimensional; 3D, three-dimensional; CCL, cathode catalyst layer; CD, current density; CFD, computational fluid dynamics; CL, catalyst layer; CPU, central processing unit; FC, fuel cell; GDL, gas diffusion layer; PEM, polymer electrolyte membrane; PEMFC, polymer electrolyte membrane fuel cell; RH, relative humidity.

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it is important to fully understand the impact of both the operating conditions and the properties of the materials such as the GDL, CL, and PEM.

The most important PEMFC operating conditions are temperature (*T*), operating pressure (*p*), relative humidity (*RH*), and stoichiometry (λ_e). Increasing temperatures rise the conductivity of the membrane and, in this way, the performance of the fuel cell. Jang et al. [1] investigated the effect of temperature on the performance of different cells in a stack. In their experimental study, they found that increasing the operation temperature of the stack augmented differences in the performance of the cells in the stack. In addition, they showed that raising the temperature of the anode led to increased humidification and, thus, to better fuel cell performance. Wasterlain et al. [2] found out that higher cell temperature and cathode stoichiometry rates elevated the electrical efficiency of the PEMFC, but indicated that this could lead to membrane drying and failure on long-term periods.

Al-Baghdadi and Al-Janabi developed a three-dimensional, nonisothermal computational fluid dynamics PEMFC model [3]. The authors applied the developed model to study the influence of operation conditions on PEMFC performance [4]. They showed that higher pressure caused a more even current density distribution at the catalyst layer. An increase in cell temperature led to a decrease in the cell activation over-potential, and a decrease in temperature caused a higher temperature gradient, resulting in a cell voltage drop. Research performed by Srinivasarao et al. [5] demonstrated similar results.

Chen et al. [6] investigated the impact of the relative humidity (RH) on fuel cell performance. A lower relative humidity prevented electrode flooding, but low RH also led to membrane drying.

Using a segmented cell, Weng et al. [7] conducted an experimental investigation of the influence of relative humidity on fuel cell efficiency with the membranes Nafion 112 and 117, each with a different thickness. Increasing the temperature and relative humidity increased fuel cell performance. The fuel cell with the thinner membrane, Nafion 112, could be applied at very low gas humidity and gas stoichiometry ratio, and displayed better performance uniformity as compared to the performance of the fuel cell with the thicker membrane, Nafion 117.

The effect of the membrane properties on PEMFC performance has been discussed by several authors [4,6,8–11]. Some [4,6,8–10] indicated that membrane thickness is an important factor, which significantly affects fuel cell performance. A thicker membrane has a higher electrical resistance and, thus, decreases the cell power density. On one hand, a thinner membrane would result in an increase in cell performance. On the other hand, a thinner membrane promotes an increase in the cross-over current, which reduces cell efficiency. Sui et al. [11] analysed the effect of the electro-osmotic coefficient on PEMFC behaviour. Decreasing the electro-osmotic coefficient resulted in a loss of the quantity of water dragged from the anode to the cathode. In this experiment, more water diffused from cathode to anode across the membrane. This resulted in an increase in performance of the fuel cell due to the improved membrane humidification on the anode side.

The influences of catalyst layer parameters such as a catalyst thickness, Pt-loading, carbon and ionomer contents, and void volume fraction have been examined in several studies [5,12–16]. Based on the results of a numerical investigation, Song et al. [12] reported that the thickness of the catalyst layer had a greater impact on cell efficiency than other parameters. An optimal catalyst thickness of around 13 μ m for the investigated fuel cell was identified. However, some slight decline of the catalyst thickness was noted to lead to a sharp drop in current density. This finding confirmed results of another investigation [13].

The impact of the GDL characteristics has been discussed by

several authors [4,17–21]. Pourmahmoud et al. [17] found that a thinner GDL had a positive effect on the mass transport and reduced the transport loss, resulting in a better cell performance. Al-Baghdadi and Al-Janabi [4] showed that an increase in GDL porosity resulted in an improvement in mass transport within the cell. The higher porosity evened out the local current density distribution. The decline in the GDL porosity could lead to hot spots. however, which would speed up dehydration and deformation of the membrane. The lower porosity elevates the transport resistance and, thus, leads to a higher water concentration gradient in the system. According to the study of Gulan and Torkoglu [18], the porosity of the gas diffusion layer had a stronger effect on cell performance than the catalyst layer porosity. They confirmed that higher GDL porosity resulted in better cell performance, confirming the results reported by Pourmahmoud et al. [17]. Using a threedimensional PEMFC model, Dawes et al. [19] determined that the GDL permeability had little to no effect on the current density, due to the diffusion-dominated nature of the gas flow.

Nabovati et al. [20] investigated the impact of the heterogeneity of the GDL porosity on its permeability and tortuosity (in- and through-plane). They found out that an increase in the heterogeneity of the porosity led to a higher level of in-plane permeability and lower in-plane tortuosity. An increase in the porosity had the opposite effect on the transport in the through-plane direction: this increase resulted in a reduction of the through-plane permeability and a growth of the through-plane tortuosity.

Using three-dimensional multiphase simulations, Jiao and Li [21] investigated the influence of both static the contact angles and sliding angles of the gas diffusion layer and catalyst layer on PEMFC performance. The results showed that increases in the sliding angle increased the liquid water volume fraction in the cathode CL and GDL several times, while changes in the static contact angle did not significantly influence the water management of the PEMFC. Since the GDL is much thicker than the CL, the alteration in the surface dynamic wettability of the GDL has more significant effect on the liquid water transport than the alteration in the surface dynamic wettability of the CL.

The current paper presents the results of a systematic computational investigation of the influence of alterations in individual physical and transport characteristics of fuel cell components such as the polymer electrolyte membrane, catalyst layer, and gas diffusion layer on the PEMFC performance.

The study was motivated by an interest to obtain an overview of the influence of material properties on the fuel cell performance. The presented results will be applied to determine most influential characteristics of the materials, which should be taken into account in a development of PEMFC degradation models. Also, the obtained results could be used for a development or modification of the materials in order to improve fuel cell performance and power density.

1.1. Object and method

The present study was carried out using the CFD code AVL FIRE. The fuel cell module of the CFD code AVL FIRE was developed in 2009 [22], and it includes a comprehensive 3D modelling approach that coupled thermal, electric, fluidic, and electrochemical phenomena [23,24]. In 2012, a newly developed membrane transport model was implemented in the module, which takes into account the dependencies of the water sorption isotherm and electroosmotic coefficient of the polymer electrolyte membrane on the temperature [25]. The main governing and auxiliary equations 8–24 of the PEMFC model utilized in the study are presented in Appendix A. The thorough test and validation of the fuel cell model was carried out on a fully coupled fuel cell with an active area of



Fig. 1. Data of the experimental validation of the PEMFC model used in the present study: a) image of the bipolar plate and end plate of the experimental fuel cell; b) computational meshes of the Full Coupled Fuel Cell, which completely corresponds to the geometry of the experimental cell; c) polarization curves at different pressure at the cathode outlet ($\lambda a/\lambda c = 1.5/2.2$; T = 70 °C; $RH_a = RH_c = 90\%$; $p_a = 101,325$ Pa); d) the distribution of the current density along the channel from inlet (y = 0 cm) to outlet (y = 12 cm)($\lambda a/\lambda c = 1.5/2.2$; T = 70 °C; $RH_a = RH_c = 90\%$; p = 101,325 Pa); e) polarization curves at different relative humidity at the cathode ($\lambda a/\lambda c = 1.5/2.2$; T = 70 °C; $RH_a = 90\%$; p = 101,325 Pa). In Figs(d–e): the measurements are presented by symbols; the simulation – by the lines. The material properties used in the validation are given in Table 2, in Column "Reference case".

25 cm². The fully coupled fuel cell is a test cell with 13 parallel straight channels. Fig. 1a demonstrates an image of the bipolar plate and end plate of the test cell with the gas channels. Fig. 1b displays the computational meshes of the full coupled fuel cell, which completely corresponds to the geometry of the experimental test cell. Fig. 1c–f presents a comparison of the simulated performance of the test cell with the experimental data. The most

important details of the experiments are given in Appendix B. As seen from the figures the calculated values are found in quite good agreement with the experimental data, confirming the validity of used PEMFC model. The polarization curves at different pressure at the cathode outlet (Fig. 1c) indicate an increase of the fuel cell current density with growing pressure. A higher pressure leads to an elevation of the oxygen concentration, resulting in higher



Fig. 2. a) The geometry of the single channel fuel cell applied in the simulations; b) 1D/3D coupling between 1D model of the membrane coated by catalyst layer and 3D Model of the gas diffusion layer.

1	Table 1
(Cell dimensions and nominal boundary conditions

Description	Value
Number of computing cells	13,992
Anode/cathode channel depth, m	$2.48 \cdot 10^{-4}$
Channel width, m	
Anode	$2.50 \cdot 10^{-4}$
Cathode	$5.00 \cdot 10^{-4}$
Cell length, m	$1.20 \cdot 10^{-1}$
Active area, m	$1.20 \cdot 10^{-1} imes 7.5 \cdot 10^{-4}$
^a Gas diffusion layer thickness, m	$3.0 \cdot 10^{-4}$
Inlets	
Mass flow via stoichiometry λ	
Anode	1.5
Cathode	2.2
Temperature, °C	70
Relative humidity	
Anode	0.90
Cathode	0.90
Outlets	
Pressure p/Pa	101,325
Channel wall	
Temperature, °C	70

^a The GDL thickness was taken from measurements at compression of 329 PSI.

reaction rates and current density. A lower inlet relative humidity (Fig. 1e) leads to a less humid membrane and, hence, to a lower proton conductivity and current density. This performance decrease occurs both in the experiment and in the simulation.

In present work, a single channel fuel cell geometry (Fig. 2a) was used in the simulation. This single channel fuel cell represents one of 13 parallel channels of the fully coupled fuel cell shown in Fig. 1b. Numerous simulations on this single channel geometry have shown that the complete cell can be represented by the single channel in a good approximation, i.e. significant macrocharacteristics of the fuel cell, such as polarization curves, total water fluxes, and over-potentials are similar for single channel geometry and fully coupled fuel cell geometry. Therefore, the current parameter study is carried out on the single channel fuel cell in order to save both computing time (about 10–15 times shorter as compared to the time needed for calculations on the full geometry cell) and CPU resources. Table 1 presents the sizes of the single channel fuel cell. The mesh contained 13 992 hexahedral computational cells. The computational mesh consisted of cathode and anode channels and gas diffusion layers. The location of the channel in- and outlets indicated a counter-flow configuration. At the surface on the left-hand side of the channels and on the right-hand side of the GDLs, symmetry boundary conditions were applied. i.e., only half of the single channel fuel cell was considered. The catalyst layers and membrane were not part of the mesh, since a separate 1D model was used in these domains to describe the transport processes across the membrane and the electrochemical reactions in the catalyst layers. This 1D model was coupled with the 3D Eulerian model in the adjacent GDL layers [23,24]. Fig. 2b shows a schematic sketch of this procedure, which take place at the interface between catalyst and gas diffusion layers. Gas species mass fraction y_i , pressure p, phase volume fractions α_g , α_h electric potential φ_s , and phase temperatures T_g , T_l , T_s in the gas diffusion layer close to the catalyst layer are used as boundary conditions for the 1D model, whereas current density *i*, water flux J_w and heat flux \dot{q} at the catalyst layer, resulting from the 1D model, act as sources for the 3D model of the GDL.

The basis for the 1D/3D coupling in *Faraday's law*, which relates the reaction mole fluxes to the current density:

$$i = N_e F \dot{n}_{s\,cl} \tag{1}$$

where N_e is the number of electrons consumed, F is the Faraday constant, and $\dot{n}_{s,cl}$ is the mole flux of the consumed gas (oxygen or hydrogen). The Faraday constant is the electric charge required for the consumption of one mole of electrons. The multiplication with N_e yields the amount of charge per consumed mole of species *s*. By taking into account Eq (1) and the electrochemical reactions at the anode (2) and cathode (3):

$$H_2 \to 2H^+ + 2e \tag{2}$$

$$0_2 + 4H^+ + 4e \to 2H_20,$$
 (3)

Table 2

List of the materials parameters and their values tested in the stu	Jdy
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	-	-				
Material		Parameter	Unit	Reference value (rv)	Variation 1 rv*1.5	Variation 2 rv*0.5
PEM	1	Thickness	μm	35	52.5	17.5
	2	Ionic conductivity	A/mV	5.37	8.05	2.68
	3	Acid group concentration	mol/m ³	1.90	2.85	0.95
	4	Water diffusion coefficient, d_w^{298}	m ² /s	$2.68 \cdot 10^{-11}$	4.02.10-11	1.34.10-11
	5	Electro-osmotic coefficient, c_{drag}^{298}	_	$1.08 \cdot 10^{-1}$	$1.62 \cdot 10 - 1$	5.41.10-2
	6	Diffusion coefficient of H_2	m ² /s	$2.09 \cdot 10^{-10}$	3.14.10-10	1.05.10-10
	7	Diffusion coefficient of O ₂	m ² /s	$9.73 \cdot 10^{-11}$	1.46.10-10	4.87.10-11
	8	Diffusion coefficient of N ₂	m ² /s	$9.73 \cdot 10^{-11}$	1.46.10-10	4.87.10-11
	9	Henry coefficient H ₂	Pa	$1.21 \cdot 10^8$	1.82.108	6.05 · 107
	10	Henry coefficient O ₂	Pa	5.14 · 10 ⁸	7.71.108	2.57.108
	11	Henry coefficient N ₂	Pa	5.14 · 10 ⁸	7.71.108	2.57.108
CL	12	Exchange current density at cathode	A/m ³	$3.2 \cdot 10^5$	4.8.105	1.6.105
	13	Exchange current density at anode	A/m ³	$1.0 \cdot 10^{11}$	1.50.1011	5.00.1010
	14	Transfer coefficient at cathode	_	0.10	0.15	0.05
	15	Transfer coefficient at anode	_	0.50	0.75	0.25
	16	Thickness of cathode	m	$1.13 \cdot 10^{-5}$	$1.69 \cdot 10 - 5$	5.63.10-6
	17	Thickness of anode	m	$1.13 \cdot 10^{-5}$	$1.69 \cdot 10 - 5$	5.63.10-6
GDL	18	Thickness	m	$3.00 \cdot 10^{-5}$	4.50.10-4	$1.50 \cdot 10 - 4$
	19	Inplane permeability	m ²	$2.33 \cdot 10^{-12}$	3.50.10-12	1.17.10-12
	20	Throughplane permeability	m ²	$1.07 \cdot 10^{-14}$	$1.61 \cdot 10 - 14$	5.35.10-15
	21	Porosity*	-	0.78	0.98	0.59
	22	Tortuosity*	-	1.50	1.88	1.13
	23	Contact angle*	Deg	122	152	92
	24	Throughplane thermal conductivity	W/mK	4.55	6.83	2.28
	25	Throughplane electrical conductivity	A/mV	$8.49 \cdot 10^2$	$1.27 \cdot 10^2$	$4.24 \cdot 10^2$

For the GDL porosity, tortuosity and contact angle the following variations were investigated: Variation 1: rv*1.25; Variation 2: rv*0.75.

the species mole fluxes at the electrodes can be formulated, yielding for the anode

$$\dot{n}_{H_{2,cl}} = -\frac{i}{2F}$$
 and $\dot{n}_{H_20,\ cl} = -J_w$ (4)

and for the cathode

$$\dot{n}_{O_{2,cl}} = -\frac{i}{4F}$$
 and $\dot{n}_{H_2O,\ cl} = \frac{i}{2F} + J_w$ (5)

Other governing equations of the applied PEMFC Model are presented at the end of the paper in Appendix A.

Table 2 displays a list of the characteristics of the polymer electrolyte membrane, catalyst layers, and gas diffusion layers, which are required input parameters to conduct the simulation of the PEMFC performance. It should be mentioned that the thicknesses of GDL, CL and PEM are related to geometry parameters of the fuel cell, which can be varied easily during the manufacture process. Other material characteristics such as the permeability, conductivity, diffusion coefficients, Henry constants, contact angle etc. are related to the material properties. The changes of these properties can be related to potential action on the material design.

All values from Table 2 correspond to the material properties at 25 °C. The magnitudes of the material parameters for the reference case are presented in the column "Reference Value". In the first step, the performance of the fuel cell was calculated for the reference case at ten different voltages ranging from 0.420 to 0.855 V.

In order to investigate the influence of each parameter on the performance of the fuel cell, the reference value of the tested parameter was increased by 50%, i.e., a variation factor of 1.5 as applied (Table 2, Variation 1), whereby the other material quantities were held unchanged at the reference values. The performance of the fuel cell was simulated at the same voltages as for the reference case. After that, the reference value of the tested parameter was decreased by 50%, i.e., a variation factor of 0.5 was applied (Table 2, Variation 2) and the fuel cell performance was simulated once again. In order to investigate the influences of the porosity, tortuosity and contact angle of the GDL, respectively, the variation factors of 0.75 and 1.25 were applied, because the factors applied for the other characteristics (1.5 and 0.5) would have led to unphysical results. In Table 2, the column labelled "Variation 1" contains the higher values of the tested parameters (the reference value multiplied by factors 1.5 or 1.25), and the column labelled "Variation 2" presents the lower values (the reference value multiplied by factor 0.5 or 0.75).

In order to determine the effect of alternations in material characteristics on PEMFC performance, the current density dependencies of the following output quantities were analysed in detail: cell voltage, power density, membrane over-potential, mean water concentration in the polymer electrolyte membrane, total water flux through the membrane, and relative humidity level at the interface PEM/CCL.

The simulations were carried out under the convenient operating conditions of the polymer electrolyte fuel cell, as displayed in Table 1. The other magnitudes and constants applied in the CFD simulations are presented in Table 3. In order to ensure proper convergence of the equations, a sufficiently large number of iteration steps (5000) is chosen. After 5000 iteration steps a steady state solution is obtained with high accuracy. The calculation time is about 30 min on 4 CPUs per simulation case. The calculations are carried out on a Precision T7600 Workstation, with Two Intel Xeon Processors E5-2667 (Six Core, 2.90 GHz Turbo, 15 MB, 8.0 GT/s), memory of 32 GB (8 \times 4GB) and frequency of 1600 MHz.

2. Results and Discussion

All obtained and analysed results from this study are presented in Appendix C. "Supplementary Data" in Figures S1–S25. Each figure in the Supplementary Data contains the results for one tested parameter. Six diagrams on the figure display the influence of alterations in the parameter on the current density dependencies of 1) the cell potential, 2) the power density, 3) the membrane overpotential, 4) the mean water concentration in the PEM, 5) the relative humidity at the interface CCL/GDL, and 6) the total water flux through the PEM.

This section discusses the most important and interesting results obtained in the study.

Fig. 3 presents the alteration in the PEMFC performance upon variations of the membrane ionic conductivity (on the left) and the membrane thickness (on the right). As seen from the figure, the increase of the membrane conductivity results in a decrease of the ohmic over-potential (Fig. 3c) and in a growth of the current density (Fig. 3a). The rise of the membrane thickness leads to an elevation of the membrane resistance (Fig. 3d) according to Ohm's Law (Eq (24)). The rising ohmic resistance results in a reduction of the cell current density (Fig. 3b). The membrane water concentration as well as the total water flux through the membrane practically does not depend on the ionic conductivity (Fig. 3e, g). But, these magnitudes depends on the polymer electrolyte thickness (Fig. 3 f, h). According to Fick's Law, used in the PEMFC Model, the water diffusive flux is inversely proportional to the membrane thickness. Thus, thinner membranes promote the water back diffusion (back diffusion - water motion toward the anode at a higher water concentration at the cathode), that provides better water distribution and increases the mean water concentration in the membrane. As seen from Fig. 3f, thicker membranes are more exposed by a drying effect with increasing current density: the mean water concentration in the membrane decreases with growing current. It should be mentioned, that the influence of the membrane thickness on the total water flux (Fig. 3h) is more pronounced at *I*>1*A*/*cm*². Gebel et al. [26] determined a water profile in running PEMFC using small-angle neutron scattering. The experimental investigation showed that the drying effect of the polymer electrolyte was stronger for higher current density. This is well correlated with the presented simulation results.

Fig. 4 demonstrates the influence of the variation of the acid group concentration on the fuel cell current density Fig. 4a, membrane over-potential Fig. 4b and membrane water concentration Fig. 4c. The alterations of the functional group concentration

Table 3

Magnitudes and constants applied in the simulations.

Magnitudes	Symbol	Value	Unit
Activation energy of electro- osmosis	E_a^{drag}	7.418 · 10 ³	J/mol
Activation energy of membrane conductivity	E_a^{σ}	9.713 · 10 ³	J/mol
Activation energy of water diffusion in the PEM	E_a^{dif}	$19.809 \cdot 10^{3}$	J/mol
Hydraulic permeability of membrane	k _d	$1 \cdot 10^{-11}$	mol/(m Pa s)
Faraday constant	F	96485.309	C/mol
Universal gas constant	R	8.31451	J/(K mol)



Fig. 3. The polarization curves (a, b), ohmic resistance (c, d), mean water concentration in the PEM (e, f) and the total water flux through the membrane (g, h) calculated for varied values of the membrane ionic conductivity (on the left) and of the membrane thickness (on the right).



Fig. 4. The polarization curves (a), ohmic resistance (b) and mean water concentration in the PEM (c) calculated for varied values of the acid group concentration in the membrane.

insignificantly effect the polarization curves and membrane overpotential (Fig. 4a), but influence the total water flux through the PEM (Fig. 4c). The acid groups are charged hydrophilic ends of the polymer, which are responsible for the water absorption. Therefore, the elevation of the concentration increases the membrane water content. According to the equation of the percolation theory (Eq. (23)), the membrane conductivity is proportional to a volume fraction of a conducting phase, which consists of the acid groups and absorbed water. Thus, increasing acid group amount rises the volume fraction of the conducting domains, resulting in the growth of the membrane conductivity and fuel cell current density.

In order to summarize and compare the large amount of simulation data obtained, the relative changes of the current density at 0.6 V (as indicated in Fig. 3a) were estimated for the

variations made in all tested material characteristics:

for Variation 1, the relative alteration of the current density at the increase of the tested parameter was calculated by:

$$\Delta I = \frac{I_{V1} - I_{rv}}{I_{rv}} \cdot 100\%, \tag{6}$$

for Variation 2, when the tested parameter decreased, the relative change of the current density was calculated by:

$$\Delta I = \frac{I_{V2} - I_{rv}}{I_{rv}} \cdot 100\%.$$
⁽⁷⁾



where I_{rv} represents the current density (CD) calculated for the reference case; I_{V1} represents the CD calculated for the increased

Fig. 5. The relative changes of the PEMFC current density at 0.6 V caused by the increase (Variation 1) and decrease (Variation 2) of the values of the single properties of the polymer electrolyte membrane.

value of the tested parameter corresponding to Variation 1; and I_{V2} , the CD computed with the decreased value of the tested parameter corresponding to Variation 2.

Fig. 5 illustrates the relative changes in the current density at 0.6 V after the increase (on the left) and the decrease (on the right) of the individual characteristics of the polymer electrolyte membrane. As seen in the figure, the variations of the Henry and Diffusion Coefficients of H₂, O₂, N₂ in the polymer electrolyte membrane lead to insignificant changes in the fuel cell current density. In addition, the investigated variations of these gas transport parameters of the membrane do not influence the magnitudes of the membrane over-potential, membrane water concentration, or total water flux through the PEM, as shown in Figs S6–11. Fig. 5 shows that the variations in the membrane thickness and specific conductivity cause the most significant changes in the current density. The decrease in the membrane thickness by 50% increases the FC current density by 26%, while the increase in the PEM thickness by 50% reduces the CD only by 17%. The reduction in the membrane conductivity by 50% yields a current density drop of 25%, while the increase in conductivity results in an increase in the current density by 13%. The increase in the water diffusion coefficient and acid group concentration raises the current density of the fuel cell by 2%, while the increase in the electro-osmotic coefficient causes the CD to decrease by 3%. The reduction in the membrane water diffusion coefficient and acid group concentration results in a drop in CD by 5%, while a decrease in the electro-osmotic coefficient elevates the current density by 3%.

The water diffusion and electro-osmotic coefficients play an important role in the water management of the PEMFC. They contribute to the total water flux, influence the relative humidity and formation of liquid water in the GDL. An accumulation of condensed water in the GDL can lead to a dramatic drop in the cell potential. Fig. 6 presents the influence of the values of the water diffusion and electro-osmotic coefficients in the membrane on the fuel cell performance. This study shows that the variations in the water diffusion and electro-osmotic coefficients investigated caused virtually no changes in the polarization curves at $0 \le I \le 1.0$ A/cm², as shown in Fig. 6a–b. At I > 1.0 A/cm² the water diffusion and electro-osmotic coefficients effect the fuel cell performance in different way: increasing water diffusion coefficient leads to a growth of the current density (Fig. 6a), while rising electro-osmotic coefficient decreases the current (Fig. 6b). As shown in Fig. 6e, with increasing water diffusion coefficient the membrane water concentration goes up. It can be explained by a fact that, a higher water diffusion coefficient accelerates the water back diffusion. It leads to an increase in the membrane water concentration and consequently to a reduction of the membrane overpotential (Fig. 6c). As contrasted to the water diffusion coefficient, growing the electroosmosis decreases the water concentration in the membrane (Fig. 6e). It happens, because the electro-osmotic flux promotes the water transport from the membrane into the gas diffusion layer, that is accompanying by the water desorption. As shown in Fig. 6h, the increase in the electro-osmotic coefficient strengthens the total water flux through the PEM. The lower water concentration in the membrane corresponds to a reduced ionic conductivity, higher ohmic over-potential (Fig. 6d) and poorer cell performance (Fig. 6b).

Fig. 7 displays the distribution of liquid water in the anode GDL at the interface of the bipolar plate/channel for the tested values of the water diffusion coefficient in the PEM. An increase in the water diffusion coefficient of the membrane leads to the formation of condensed water, whereby the volume fraction of the liquid water grows in locations that are removed from the gas channel.

Fig. 8 illustrates the distribution of the liquid water in the

anode at the interface of the GDL/bipolar plate for the tested values of the electro-osmotic coefficient. The increase in electro-osmotic coefficient reduces the concentration of condensed water in the anode.

Fig. 9 shows the relative changes in the current density at 0.6 V caused by variations in catalyst layers parameters. As seen from the figure, the alterations in characteristics of the anode catalyst layer do not lead to any visible changes in the current density of the fuel cell. Also, no changes are observed in the other analysed quantities such as the membrane over-potential, mean water concentration, relative humidity, and water flux shown in Figures S12–S17. Increasing the thickness and exchange current density of the cathode catalyst layer result in an increase in the cell CD of around by 7%, while increasing the transfer coefficient at the cathode decreases the CD by 7%. The influences of variations in the exchange current density and transfer coefficient of the cathode on the polarization curves are shown in Fig. 10.

Fig. 11 displays the relative changes of fuel cell current density upon variations in the GDL characteristics. Significant effects are observable as changes in the electrical conductivity, thickness, porosity, and contact angle of the GDL.

In paper [27], Kulkarni and Wang presented a sensitivity analysis of some gas diffusion layer parameters on PEMFC performance. The study was performed using a non-isothermal 2D fuel cell model and COMSOL Multiphysics. The analysis showed that the porosity and through-plane permeability were the most influential GDL parameters in the FC performance. The experimental studies [28,29] demonstrated that the transport characteristics and surface properties of GDLs can be varied using different polytetrafluoroethylene (PTFE) loading. It was shown that the contact angle of GDL increases with increasing PTFE content, but the gas permeability and through-plane electrical conductivity decrease at a higher PTFE loading. Additionally, Chun et al. [29] modelled the FC performance as changes in the GDL thickness and permeability. Lowing fuel cell performance was reported for thicker GDLs. The simulation showed that the GDL's gas permeability increased by three orders of the magnitude led to a light increase of the FC voltage. In experiment measurements [29], a significant effect on the FC performance was detected upon an alteration of the through-plane conductivity of the GDL: decreasing GDL conductivity caused lowing cell voltage. These simulation and experimental results are found in a good agreement with the present calculations.

The alteration of the GDL thickness leads to a change of the distribution and concentration of the gases and water near the catalyst. With increasing GDL thickness, the liquid water saturation rises rapidly, but the oxygen concentration near the catalyst layer decreases. In addition, as through-plane electrical resistance increases with increasing GDL thickness, thicker GDL causes decline in the cell performance.

The present simulation demonstrates a very interesting result for the porosity. As seen from Fig. 11, increasing the GDL porosity by 25% leads to a dramatic drop in the current density of around by 70%, while a reducing this parameter by 25% elevates the current density by only 9%. The increase in the GDL porosity from 0.780 to 0.975 causes a very rapid transport of gases and water through the GDL in an in-plane direction. In this way, water and gases are quickly transported from the interface of the polymer electrolyte membrane to the gas channels of the bipolar plate. This leads to a drop in the relative humidity (Fig. 12a) at the interface PEM/GDL and, consequently, the mean water concentration in the membrane decreases (Fig. 12b). The lower water content in the PEM increases the membrane ohmic resistance (Fig. 12c) and, in this way, decreases the current density (Fig. 12d).



Fig. 6. The polarization curves (a, b), ohmic resistance (c, d), mean water concentration in the PEM (e, f) and total water flux through the membrane (g, h) calculated for varied values of the water diffusion coefficient (on the left) and electroosmotic coefficient (on the right) in the membrane.

3. Conclusion

The paper summarized the results of a systematic computational investigation of the effects of alterations in characteristics of the polymer electrolyte membrane, catalyst layers, and gas diffusion layer on the performance of a polymer electrolyte fuel cell. The influences of 25 parameters were tested on a single channel fuel cell using the CFD code AVL FIRE:



Fig. 7. The distribution of the liquid water in the anode GDL at the interface to the bipolar plate/channel at different values of the water diffusion coefficient in the PEM, $d_{2}^{998}[m^2/s]$: a)1.34·10⁻¹¹(V2); b)2.68·10⁻¹¹(rV); c) 4.02·10⁻¹¹(V1).



Fig. 8. The distribution of the liquid water in the anode GDL at the interface to the bipolar plate/channel at the different values of the electro-osmotic coefficient in the PEM, c_{dreg}^{298} : a) 0.054(V2); b) 0.108(rv); c) 0.160(V1).



Fig. 9. The relative changes of the PEMFC current density at 0.6 V caused by the increase (Variation 1) and decrease (Variation 2) of the values of the single properties of the catalyst layers.



Fig. 10. Polarization curves calculated for the different values of the cathode catalyst layer: a) exchange current density; b) transfer coefficient.



Fig. 11. The relative changes of the PEMFC current density at 0.6 V caused by the increase (Variation 1) and decrease (Variation 2) of the values of the single properties of the gas diffusion layer. For Variation 1, the result for the GDL porosity is related to the right axis.

- PEM properties: the thickness, acid group concentration, ionic conductivity, electro-osmotic coefficient, water diffusion coefficient, Henry coefficients as well as the diffusion coefficients of H₂, O₂, N₂;
- CL parameters at the cathode and anode: the thickness, exchange current density, transfer coefficient; and
- GDL characteristics: the thickness, porosity, in-plane permeability, through-plane permeability, tortuosity, contact angle, thermal conductivity, and through-plane electrical conductivity.

The simulations for the PEMFC performance were conducted by increasing and decreasing the values of each tested parameter and comparing the results to those of the reference case. In order to analyse changes in PEMFC performance, the current density dependencies of the following output quantities were investigated in detail: 1) the cell potential, 2) the power density, 3) the membrane over-potential, 4) the mean water concentration in the PEM, 5) the relative humidity at the interface CCL/GDL, and 6) the total water flux through the PEM.



Fig. 12. The current density dependencies of the fuel cell characteristics calculated at the different values of the GDL porosity: a) the cell voltage; b) the membrane overpotential; c) the mean water concentration in the PEM; d) the relative humidity at the interface the CCL/PEM.

Table 4
The parameters influencing the current density (at $U=0.6~V$) by more than 5%

Materials parameters	Variation 1: rv*1.5	Variation 2: rv*0.5
PEM		
Thickness	-17%	+26%
Conductivity	+13%	-25%
Water diffusion coefficient	+2%	-6%
Acid group concentration	+2%	-6%
CL		
Cathode thickness	+7%	-12%
Cathode exchange current density	+7%	-12%
Cathode transfer coefficient	-7%	+7%
GDL		
Electrical conductivity	+13%	-25%
Thickness	-14%	+18%
Porosity*	-70%	+9%
Contact angle*	+1%	-7%

For the GDL porosity, and contact angle the following variations were investigated: Variation 1: rv*1.25; Variation 2: rv*0.75.

The material parameters, changes in which caused corresponding changes in the current density (estimated for 0.6 V) of the PEMFC by more than 5%, are presented in Table 4. Alterations in the conductivities and thicknesses of the PEM and GDL as well as GDL porosity led to more significant changes in the fuel cell performance than alterations in other material properties. Alterations in the values of the characteristics of the anode catalyst layer tested showed little influence on the fuel cell behaviour. The increase in the thickness and exchange current density of the cathode catalyst layer elevated the fuel cell current density, while the increment of the transfer coefficient decreased the fuel cell performance.

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Appendix A. Governing equations of the PEMFC Model applied in the study

Cell potential :
$$U_{cell} = U_{oc} - \eta_c - \eta_a - \eta_{mem}$$
 (8)

Mass and momentum balance in the gas channels and GDLs

The GDL is treated as a porous solid phase *s* in the *Eulerian multiphase approach*.

Species *i* mass in gas phase *g*:

$$\nabla \cdot \left(\alpha_g \rho_g y_i \overrightarrow{u}_g \right) = -\nabla \cdot \left(a_g \overrightarrow{j}_i \right) + \dot{M}_{pc,i} + \dot{M}_{rl,i} \quad \text{for } i = 1, \dots, N_e$$
(9)

Phase
$$\boldsymbol{p}$$
 mass $\nabla \cdot (\alpha_p \rho_p \overrightarrow{u}_p) = \dot{M}_{pc,c} + \dot{M}_{rl,p}$ for $p = g, l$ (10)

Phase **p** momentum $\nabla \cdot (\alpha_p \rho_p \overrightarrow{u}_p \overrightarrow{u}_p)$

$$= -\alpha_p \nabla p + \nabla \cdot (\alpha_p \tau_p) + S_p + S_{pc,p} \text{ for } p = g, l$$
(11)

Phase **p** total enthalpy $\nabla \cdot (\alpha_p \rho_p H_p \overrightarrow{u}_p)$

$$= \nabla \cdot (\alpha_p \kappa_p \nabla T_p) + \nabla \cdot (\alpha_p \tau_p \cdot \vec{u}_p) + \dot{Q}_p + \dot{Q}_{D,p} + \dot{Q}_{pc,p} \text{ for } p = g, l$$
(12)

$$\mathbf{0} = \nabla \cdot \left(\alpha_{s} \kappa_{s} \nabla T_{p} \right) + \dot{Q}_{s} + \dot{Q}_{rl} + \dot{Q}_{\sigma}$$
(13)

Electric charge in solid phase $\mathbf{s} - \nabla \cdot (\sigma_s \nabla \phi_s) = R_{cl}$ (14)

Electrochemical reactions

The relationship of the current density with the electrode overpotentials is described by a Butler–Volmer equation,

for the cathode reaction :
$$i = i_{0,c} l_{cl,c} \left[-exp \left(-\frac{k_c F}{RT_m} \eta_c \right) + \frac{c_{O_2,sf}}{c_{O_2,ch}} exp \left(\frac{(1-k_c)F}{RT_m} \eta_c \right) \right]$$
(15)

for the anode reaction :
$$i = i_{0,a} l_{cl,a} \left[\frac{\sqrt{c_{H_2,sf}}}{\sqrt{c_{H_2,ch}}} exp\left(\frac{k_a F}{RT_m} \eta_a\right) - exp\left(-\frac{(1-k_a)F}{RT_m} \eta_a\right) \right]$$
(16)

Membrane transport model

Total water flux through the membrane :

$$J_w = -a \cdot D_w \frac{dC_w}{dz} + C_{drag} \cdot \frac{i}{F} - k_d \frac{dp}{dz}$$
(17)

Boundary conditions of water transport,

at the anode :
$$-a \cdot D_{w} \frac{dC_{w}}{dz} + C_{drag} \frac{i}{F} - k_{d} \frac{dp}{dz}$$
$$= -a \cdot \gamma_{H_{2}O,a} \left(C_{w}^{a} - C_{w}^{a^{*}}\right)$$
(18)

at the cathode :
$$-a \cdot D_w \frac{dC_w}{dz} + C_{drag} \frac{i}{F} - k_d \frac{dp}{dz} + \frac{i}{2F}$$
$$= a \cdot \gamma_{H_2O,c} \left(C_w^c - C_w^{c*} \right)$$
(19)

Water sorption isotherm :

$$C_{w}^{*} = \left(1.55 + 13.71 \cdot \varphi - 24.37 \cdot \varphi^{2} + 21.87 \cdot \varphi^{3}\right) * f(c_{w}^{\max}, \varphi)$$
(20)

where $f(c_w^{\max}, \varphi)$ – is a multiplier, which is directly proportional to the maximal membrane water concentration c_w^{\max} described by $c_w^{\max} = 0.138 T - 28.31$ and the relative humidity φ .

Water diffusion coefficient :

$$D_w^{T_2} = d_w^{T_1} \cdot C_w \cdot \exp\left[-\frac{E_a^{dif}}{R} \cdot \left(\frac{1}{T_2} - \frac{1}{T_1}\right)\right]$$
(21)

Electro - osmotic coefficient :

$$C_{drag}^{T_2} = c_{drag}^{T_1} \cdot C_W \cdot \exp\left[-\frac{E_a^{drag}}{R} \cdot \left(\frac{1}{T_2} - \frac{1}{T_1}\right)\right]$$
(22)

Membrane conductivity :

$$\sigma_{mem}^{T_2} = \sigma^{T_1} \cdot (f - f_{cr})^t \cdot \exp\left[-\frac{E_a^{\sigma}}{R} \cdot \left(\frac{1}{T_2} - \frac{1}{T_1}\right)\right]$$
(23)

The membrane over

 $\eta_{mem} = \frac{i}{\sigma_{mem}} \cdot L_{mem}$

Appendix B. Brief description of the experiments on the validation of the PEMFC model

The experimental measurements were carried out in the Christian Doppler (CD) Laboratory for Fuel Cell Systems at TU Graz using a fully automated test rig [30–32]. The test rig is capable of supplying well defined mixtures of hydrogen, nitrogen, oxygen, and air by using mass flow controllers. Bubble humidifiers are utilised for gas humidification, and the humidity levels are checked with downstream humidity sensors. Electrochemical potentials are measured utilising a USB multiflexer card from National Instruments (NI USB-6218). The fuel cell is composed of carbon bipolar plates with 13 straight parallel flow channels (Fig. 1a). The cathode bipolar plate is segmented into 10 individual elements, which are electronically insulated from each other. The bipolar plates are embedded between two end plates made of stainless steel. Cooling channels are installed in the end plates, which provide a precise temperature control of the fuel cell by applying water as the cooling fluid. The active area of the single fuel cell is 25 cm^2 .

Nomenclature

Latin symbols

а	sulfonic acid group concentration mol/m ³
С	normalized water concentration
C_w	normalized water concentration in the membrane at
	current condition
c_w^{\max}	normalized water concentration in the membrane,
	equilibrated with liquid water
C _{drag}	electro-osmotic coefficient (drag coefficient) in
0	membrane
$c_{drag}^{T_1}$	electro-osmotic coefficient in membrane at $C_w = 1$ and T_1
D_w	water diffusion coefficient in membrane [m ² /s]
$d_w^{T_1}$	water diffusion coefficient in membrane at $C_w = 1$ and T_1
	$[m^2/s]$
Ea	activation energy [J/mol]
F	Faraday constant [(A·s)/mol]
f	volume fraction of the conducting phase in the
-	membrane
fcr	critical volume fraction of the conducting phase

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i, *I* electric current density [A/m²]

- i_0 exchange current density [A/m³]
- \vec{j} diffusive mass flux [kg/(m²s)]
- J_w water flux [mol/(m²s)]
- *k* electrode transfer coefficient
- k_d hydraulic permeability of the membrane [mol/(m .Pa .s)]
- *l*_{cl} thickness of the catalyst layer [m]
- *L_{mem}* membrane thickness [m]
- \dot{M} volumetric mass transfer rate [kg/(m³s)]
- \dot{n} mole flux [mol/(m²s)]
- *N_e* number of gas species in electrode *e*
- *p* pressure [Pa]
- \dot{q} heat flux [J/(m²s)]
- \dot{Q} volumetric heat transfer rate [J/(m³s)]
- *R* universal gas constant [J/(mol. K)]
- R_{cl} volumetric transfer current [A/m³]
- \overrightarrow{S} volumetric transfer force [N/m³]
- T temperature [K]
- *t* exponent of the percolation theory equation
- \vec{u} velocity [m/s]
- U potential [V]
- *y_i* gas species mass fraction
- z normal direction membrane [m]

Greek symbols

α volume fraction

φ	water activity in gas phase, relative humidity
, ΥΗ ₂ Ο	water transfer coefficient [m/s]
η	potential loss [V]
κ	thermal conductivity [J/(m·K·s)
λ	stoichiometric flow ratio
ρ	mass density [kg/m ³]
σ	pre-factor of the percolation theory equation [S/m]
σ_{mem}	specific membrane conductivity [S/m]
σς	electrical conductivity of solid phase [S/m]

- τ tortuosity
- ϕ electric potential [V]
- *φ* relative humidity

Subscripts and superscripts

- *a* anode
- *c* cathode
- ch channel
- *cl* related to catalyst layer
- *drag* related to drag transport of water
- g gas phase
- *i* species index
- *l* liquid water
- *m; mem* membrane
- oc open circuit
- p phase index
- *pc* phase change
- s solid phase
- *sf* electrode surface
- w; H_2O water

Appendix C. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.jpowsour.2015.07.011.

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