

COMPUTATIONAL FLUID DYNAMICS CALCULATION OF A PLANAR SOLID OXIDE FUEL CELL DESIGN RUNNING ON SYNGAS

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The present study deals with modelling and validation of a planar Solid Oxide Fuel Cell (SOFC) design fuelled by gas mixture of partially pre-reformed methane. A 3D model was developed using the ANSYS Fluent Computational Fluid Dynamics (CFD) tool that was supported by an additional Fuel Cell Tools module. The governing equations for momentum, heat, gas species, ion and electron transport were implemented and coupled to kinetics describing the electrochemical and reforming reactions. In the model, the Water Gas Shift reaction in a porous anode layer was included. Electrochemical oxidation of hydrogen and carbon monoxide fuels were both considered. The developed model enabled to predict the distributions of temperature, current density and gas flow in the fuel cell.

Keywords: planar Solid Oxide Fuel Cells, Computational Fluid Dynamics, syngas

1. INTRODUCTION

Solid Oxide Fuel Cells, in contrast to PEMFCs, can be fed directly with hydrocarbon fuels containing methane, carbon monoxide or even carbon dioxide generated by reformers (Barelli et al. 2017; D'Andrea et al., 2017; Kang et al. 2008; Stoeckl et al., 2017). No need to eliminate CO through catalytic reactors makes that as a main advantage of using SOFCs. In addition, according to Park et al. (2012) an overall electrochemical reaction rate in the SOFC can vary up to 50% with the oxidation of CO in comparison to only oxidation of hydrogen. Therefore, both hydrogen oxidation and carbon monoxide oxidation should be included in a cell level study. Anode species composition exhibits notable amounts of CO (Tweedie and Lemcoff, 2014). Carbon monoxide oxidation was considered at the cell level modelling by Andersson et al. (2013), Ho et al. (2009), Iwai et al. (2011) as well as Razbani et al. (2013) and at the stack level by Gholaminezhad et al. (2017) and Papurello et al. (2017). Iwai et al. (2011) developed a numerical model for an anode supported, intermediate temperature direct internal reforming planar SOFC. In the simulations steam reforming reactions using methane, the Water-Gas-Shift (WGS) reaction and the electrochemical reactions of hydrogen and carbon monoxide were taken into account. Results showed that the endothermic steam reforming reaction led to a reduction in the local temperature near the inlet and limited the electrochemical reaction rates therein. However, it was found that the local temperature and current density distributions can be controlled by tuning the pre-reforming rate (Iwai et al., 2011). A multi-physics mathematical channel-level direct internal reforming methane fed Solid Oxide Fuel Cell model was presented by Gholaminezhad et al. (2017). The proposed model considered steady-state multi-component mass transport in fuel channel and its coupled effects on electrochemical phenomenon inside the anode porous electrode. Authors

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studied methane conversion, chemical reaction rates, inlet composition and carbon formation boundary as a function of pre-reforming rate, fuel utilisation, current density and temperature. However, Gholaminezhad et al. (2017) ignored in their study the electrochemical oxidation of CO due to the fact that the rate of CO conversion via Water Gas Shift, WGS, reaction was much higher than that with electrochemical oxidation of CO. Authors explained that due to the existence of a high amount of steam in the inlet stream, because of carbon deposition prevention, steam reforming was considered as the only reaction, in which methane was involved. Exclusion of CO from the electrochemical model had generated less accurate results.

Direct internal methane reforming in the high temperature SOFC was investigated by Ho et al. (2009) using Star-CD with in-house developed subroutines. Carbon monoxide produced by the reforming reaction was included in the electrochemical processes, while being in equilibrium with the WGS reaction. Co- and counter flow configurations were tested. For co-flow a sub-cooling effect manifests itself in the methane rich region near the fuel entrance, while for counter flow a super heating effect was noticed in the downstream, where all the methane was consumed.

A fully coupled 3D model was applied also by Razbani et al. (2013) to a planar electrolyte supported SOFC fed by methane free biogas using the COMSOL software. Simulation results showed that using the methane free biogas a more uniform current density profile was obtained due to the high CO₂ content and the reverse WGS reaction. It was noticed that the WGS reaction was faster than the electrochemical reactions. Razbani et al. (2013) explained this behaviour due to the endothermic reverse WGS reaction, when CO and H₂O were produced and caused fast consumption of H₂ and CO₂. The contribution of CO oxidation in the overall current productions ranges from 7% to 17% for cell voltage from 0.9 V to 0.7 V. Andersson et al. (2013) showed that a fuel mixture containing a high fraction of electrochemical reactants such as hydrogen and carbon monoxide enables a high Nernst potential in the region close to the inlet, which increases the current density there. Thus, the Nernst potential covering the electrochemical reaction with carbon monoxide had a stronger influence by changes in the operating temperature in comparison to the reaction with hydrogen only. This relationship will vary for different cell design and operating conditions when the fuel cell was designed. Therefore, the aim of this study was to apply an earlier developed three-dimensional numerical model for pure hydrogen (Pianko-Oprych et al., 2016) to a planar Solid Oxide Fuel Cell design fuelled by reformat mixtures. The idea was to check whether the model after adjustment for the presence of carbon monoxide will be able to predict thermal and electrochemical fuel cell performance. In order to visualise electrochemical and gas phase reaction regions the species and current density distributions were displayed. The obtained results will be used as guidelines for further investigations and identification of proper flow paths that increase the SOFC performance.

2. PHYSICAL AND NUMERICAL MODEL

In the SOFC, electrical and ionic conduction, heat transfer, gas phase mass transport as well as chemical reactions take place simultaneously. To include all the phenomena, conservation equations for momentum, mass, species and energy were applied to the SOFC domains. The Navier-Stokes equations were used to model the flow in the anode and cathode flow channels, while the Brinkman equations were utilised to model the flow in the porous electrodes. For the mass balances, the concentrated species transport equations were used assuming that species transport was dominated by the gas diffusion. Thermal energy was transferred by conduction and convection, while the radiative heat transfer was neglected due to its low impact (Pianko-Oprych et al., 2014). For the electronic and ionic charge balance the appropriate distributed charge transfer equations were applied. The electron transport was considered in order to collect current from the fuel cell. More details on the applied model can be found in (Pianko-Oprych et al., 2016).

In this study, the following electrochemical oxidation reactions of H₂ and CO were considered:



Carbon monoxide can be oxidized in the electrochemical reaction (Eq. (2)), but can also react with water (Eq. (4)) in the Water Gas Shift (WGS) reaction:



The cell voltage was determined by setting boundary conditions such that the electrical potential, $\phi_{\text{cell}} = 0$ in the anode side, while $\phi_{\text{cell}} = E_{\text{cell}}$ in the cathode side (current tap surface). For the study, the cell voltage was varied from 1.1 to 0.3. To calculate the volumetric current densities, the Butler-Volmer equations were used (Eqs.(5-7)):

$$i_{\text{H}_2} = i_{0,\text{an},\text{H}_2} \left[\exp\left(\frac{\alpha_{\text{anode}} n \eta_{\text{act},\text{an},\text{H}_2} F}{RT}\right) - \exp\left(\frac{\alpha_{\text{cathode}} n \eta_{\text{act},\text{an},\text{H}_2} F}{RT}\right) \right] \quad (5)$$

$$i_{\text{CO}} = i_{0,\text{an},\text{CO}} \left[\exp\left(\frac{\alpha_{\text{anode}} n \eta_{\text{act},\text{an},\text{CO}} F}{RT}\right) - \exp\left(\frac{\alpha_{\text{cathode}} n \eta_{\text{act},\text{an},\text{CO}} F}{RT}\right) \right] \quad (6)$$

$$i_{\text{O}_2} = i_{0,\text{cat},\text{O}_2} \left[\exp\left(\frac{\alpha_{\text{anode}} n \eta_{\text{act},\text{an},\text{O}_2} F}{RT}\right) - \exp\left(\frac{\alpha_{\text{cathode}} n \eta_{\text{act},\text{an},\text{O}_2} F}{RT}\right) \right] \quad (7)$$

where: $i_{\text{H}_2}, i_{\text{CO}}, i_{\text{O}_2}$ are the current density, $i_{0,\text{an},\text{H}_2}, i_{0,\text{an},\text{CO}}, i_{0,\text{cat},\text{O}_2}$, are the effective exchange current density, F is the Faraday's constant, n is the number of electrons that are released per reaction, R is the universal gas constant, T is the absolute temperature and α_{anode} and α_{cathode} are anodic and cathodic transfer coefficients, respectively.

The computational domain of a planar SOFC design consisted of three ceramic layers of membrane electrode assembly: anode, electrolyte, cathode and of two cross-flow bipolar plates with 26 ribs as shown in Fig. 1a. The gases flowed in part horizontally or vertically and diagonally in average. The fuel and air flows were cross-wise opposed on each bipolar plate side.

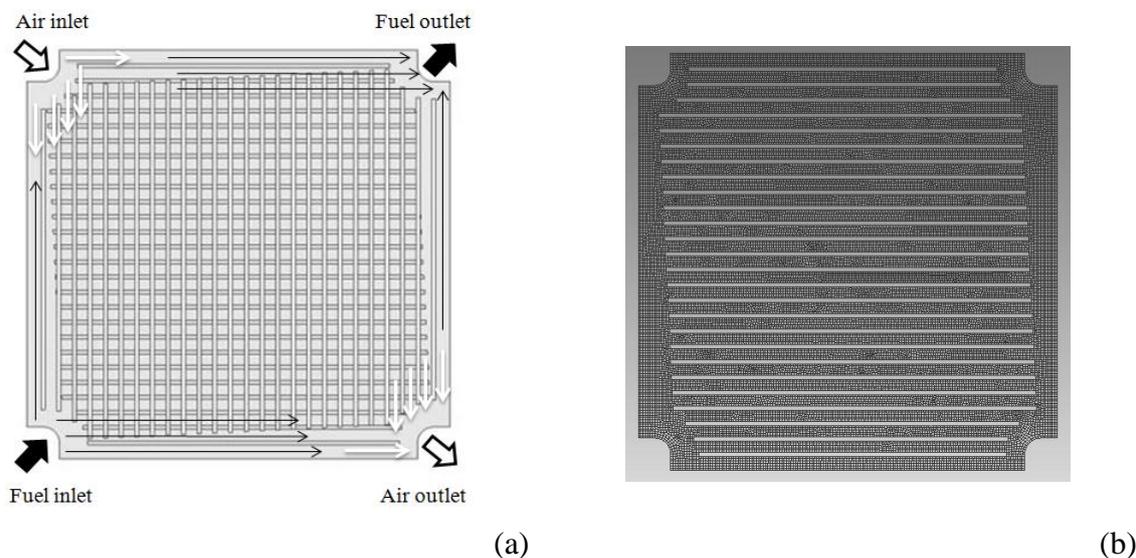


Fig. 1. A schematic of the planar Solid Oxide Fuel Cell design (a) and numerical mesh (b)

The mesh consisted of 890 thousand computational cells generated in the ANSYS Meshing 15.0 and it is presented in Fig. 1b. The physical properties of the SOFC are shown in Table 1, where YSZ means Yttria-Stabilized Zirconia, Ni - Nickel, LSM - Lanthanum Strontium Manganite. The operating conditions and input parameters for the SOFC model are listed in Table 2. All layers of the SOFC membrane-electrode assembly including a thin cathode were resolved in the model with one exception – a thin electrolyte was considered as electrode-electrolyte interfaces.

Table 1. Physical properties of the SOFC

Parameter	Anode	Electrolyte	Cathode	Current collectors
Material	Ni-YSZ	YSZ	LSM	-
Thickness, mm	0.25	0.01	0.06	1
Density, kg/m ³	7740	6000	5300	7450
Specified heat capacity, J/(kg·K)	595	400	607	600
Thermal conductivity, W/(m·K)	6.23	2.7	10	27
Resistivity, Ohm·m	-	0.1	-	-
Electronic conductivity, 1/(Ohm·m)	30300	-	12800	769000
Anode-current collectors contact resistance, Ohm·m ²	1×10 ⁻⁷	-	1×10 ⁻⁸	-
Porosity	0.3	-	0.3	-

Table 2. Boundary conditions of the SOFC model

Domain region	Parameter	Value
Air inlet	Flow rate:	0.25 l/min
	Temperature:	973 K
	Mass fraction of species:	23.3% O ₂ , 76.7% N ₂
Fuel inlet	Flow rate:	0.075 l/min
	Temperature:	973 K
	Mass fraction of species:	75% H ₂ , 25% CO
Air outlet	Pressure:	1×10 ⁵ Pa
Outer current collector (anode side) surface	Voltage tap surface:	0 V
Outer current collector (cathode side) surface	Current tap surface:	0.7 V
Anode	Exchange current density	7460 A·m ²
Cathode	Exchange current density	10090 A·m ²

In order to attain convergence for the momentum, energy and species differential equations the second order upwind discretisation scheme was used, while for the pressure equation the second order discretisation scheme was applied. Discretisation of the electric potential by the first order upwind scheme was considered, while for gradient estimations the Green Gauss node was used. The 3D computational model was developed in the commercial CFD code ANSYS Fluent 15.0 with an additional ANSYS Fuel Cell Tools module.

3. RESULTS AND DISCUSSION

The current - voltage curve was constructed and validated against the experimental data of Bossel (2015). A comparison of the calculated voltage vs. current curve with the experimental results for syngas is presented in Fig. 2. The CFD curve corresponds quite well with the experimental one for the operating voltage of 1.1 and 0.7 V, while at the lowest analysed voltage of 0.3 V significant deviation in the electrical current was noticed for the reformate. A sudden drop of power was observed at the highest current value and it can be explained by fuel starvation. Similar behaviour was observed in the previous paper (Pianko-Oprych et al., 2016), when pure hydrogen was used as a working fuel. This type of behaviour may result from the applied numerical procedure, in which the flow rates of the fuel and air for considered voltage values have to be kept constant, while during measurements the flow rate is adjusted according to the working conditions inside the fuel cell in order to avoid fuel cell destruction due to reagent starvation.

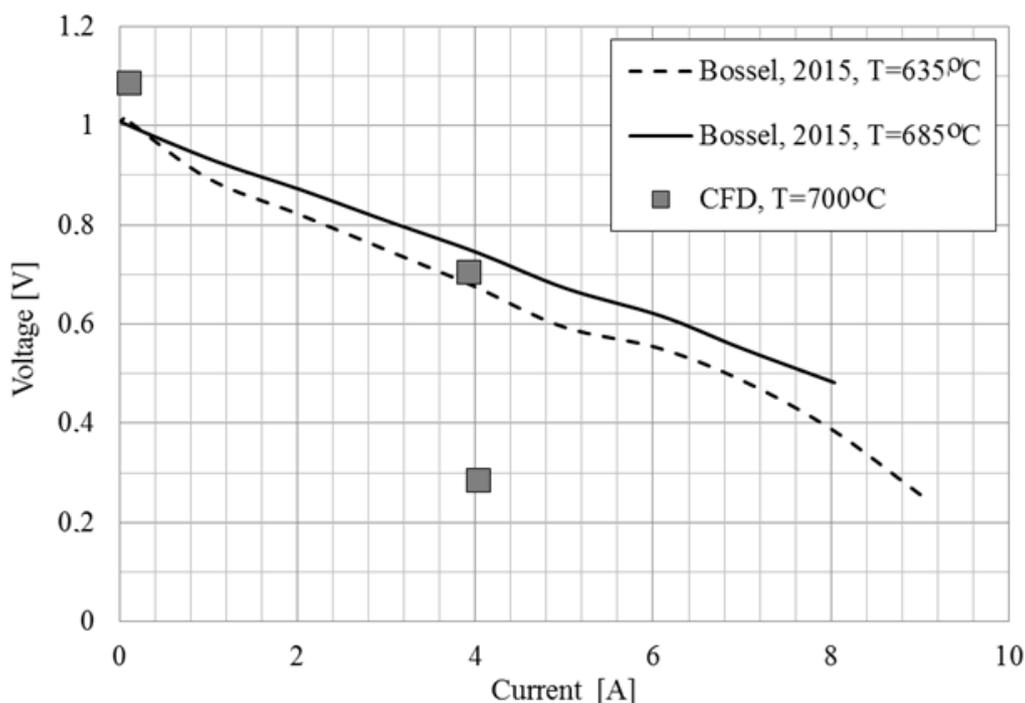
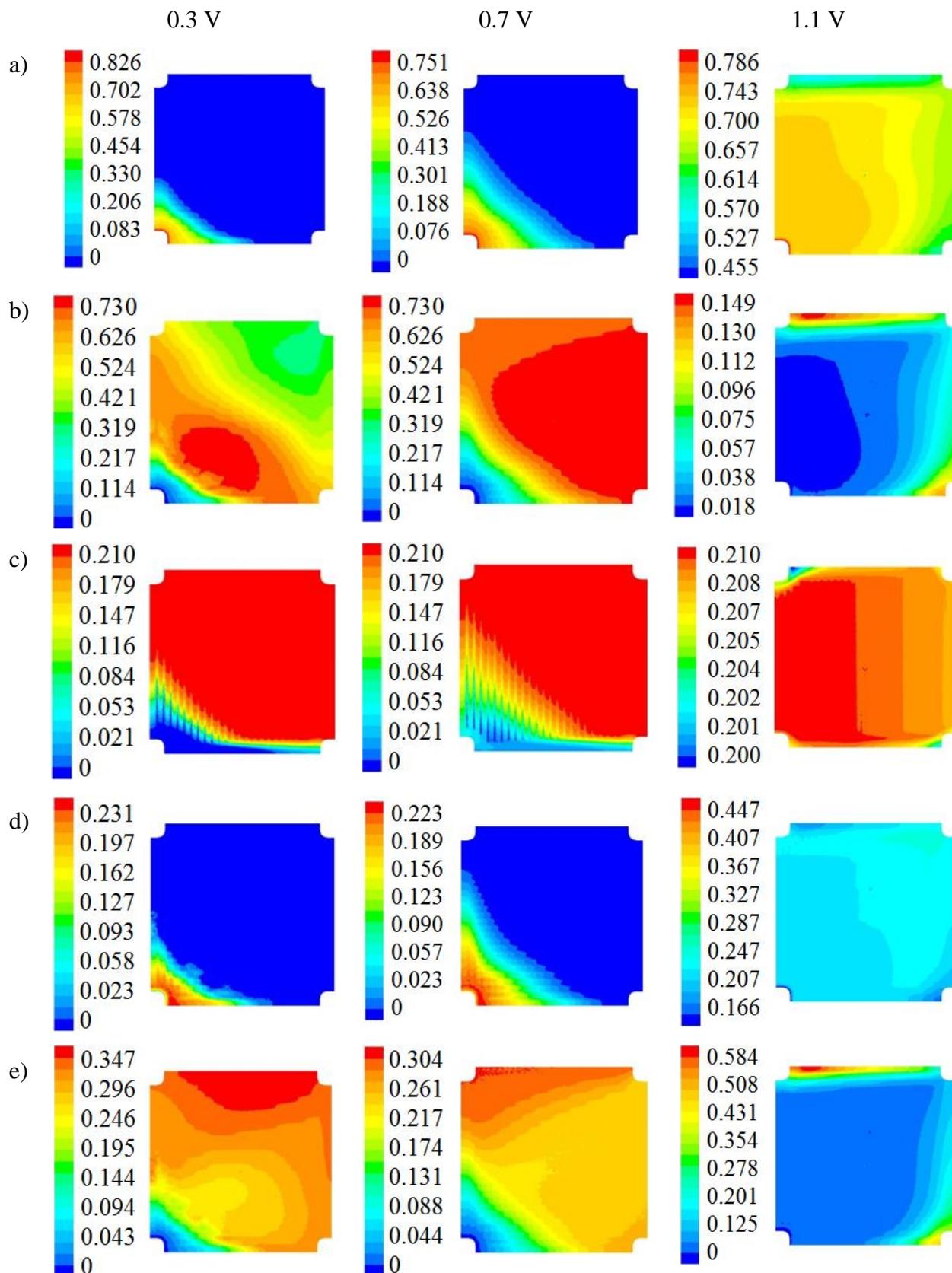


Fig. 2. Voltage [V] vs. current [A] curve for the single planar SOFC at temperature of 700 °C and fed by syngas

An attempt to explain such SOFC behaviour was undertaken. For this purpose, hydrogen, water, oxygen, CO and CO₂ mass fraction distributions were shown in Table 3, respectively. The fuel inlet of hydrogen and CO was located in the left bottom corner, while the air inlet was located in the upper left corner. The mean fuel and air flow directions were across the fuel cell and perpendicular to each other. The mass fractions of hydrogen and carbon monoxide significantly change diagonally between the fuel inlet and outlet. The simulation results indicate the regions where hydrogen and carbon monoxide were available electrochemically highly active and the fuel cell was locally able to produce current at the applied voltage, while in the areas where local fuel starvation can be noticed, the cell could not produce enough electrical current particularly at the cell voltage of 0.3 V. It can be noticed that the areas where hydrogen and carbon monoxide concentrations strongly decreased (Tables 3a and 3d) overlap with regions of the highest current density (Table 5). What was surprising the WGS reaction showed the most activity at the upper part of the cell in the regions close to the air inlet and fuel outlet surfaces, where a higher amount of CO₂ was noticed at the electrolyte interface from the anode side. On the other hand, increasing the mass fraction of water decreased the probability of carbon formation although temperature gradients across the fuel cell may contribute to its occurrence.

Table 3. Table 3. Species distributions [kg/kg] in the electrolyte from the anode side: (a) mass fraction of hydrogen, (b) mass fraction of water, (c) mass fraction of oxygen, (d) mass fraction of CO, (e) mass fraction of CO₂



Therefore, the key parameter in the electrochemical analysis of the planar SOFC is temperature. Temperature distribution at the electrolyte from the anode side fuelled by syngas is shown in Table 4. The highest temperature was noticed at the lowest operating cell voltage of 0.3 V. This is likely due to the high concentrations of hydrogen and carbon monoxide at the inlet, which result in an increase in the electrochemical heat generation. Temperature distributions were highly non-uniform in the planar fuel cell and they differed quite significantly from each other for the operating voltage range values. Indication of hot spots was critical for the fuel cell safety reasons, because too high temperature gradients within the cell can lead to its damage.

The consumption rates of hydrogen, CO and oxygen as well as the formation rates of water and CO₂ affect the current density distributions presented in Table 5. Areas of the highest current density appear in the same regions where the highest formation rates were noticed.

Table 4. Distributions of temperature [K] at the electrolyte from the anode side of SOFC fed by syngas

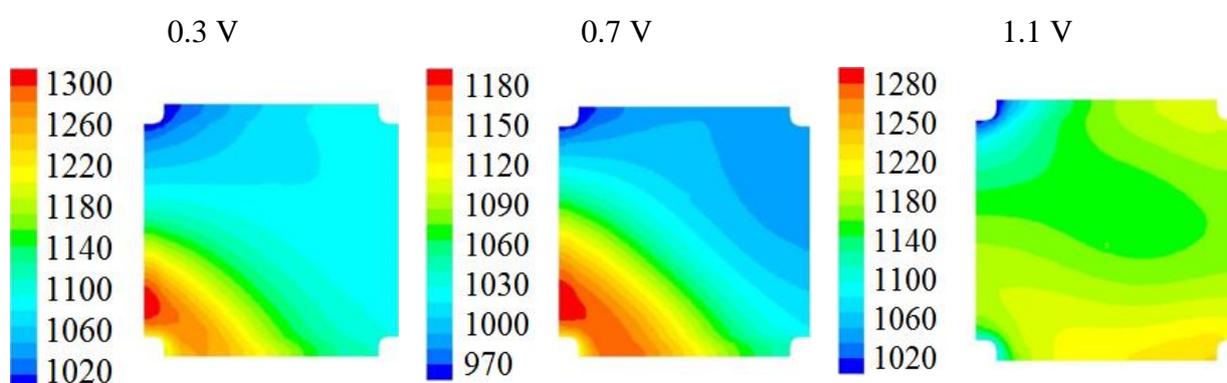
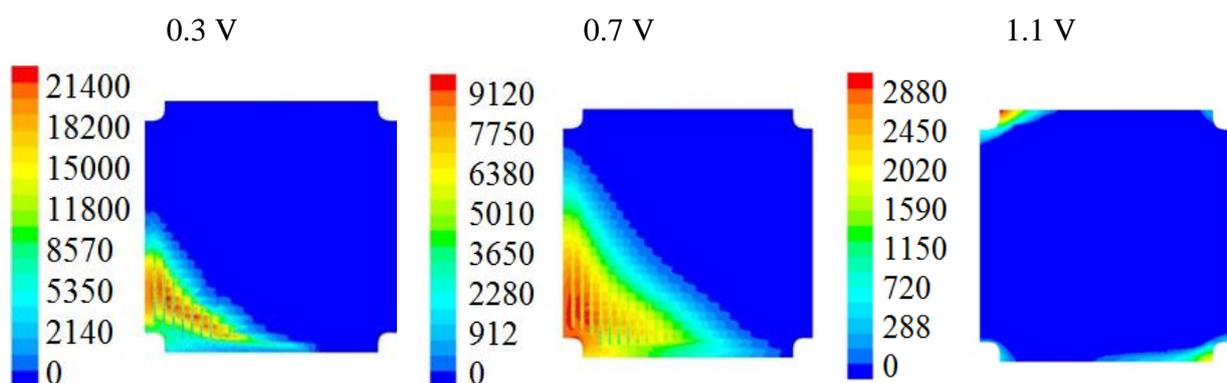


Table 5. Current density distributions [A/m²] at the electrolyte from the cathode side of SOFC fed by syngas



4. CONCLUSIONS

The presented modelling investigation was carried out to estimate the performance of the planar SOFC fuelled by a mixture of hydrogen and carbon monoxide and to provide a forecasting tool to predict fuel cell performance in relation to fuel composition changes. It was quantitatively shown that the fuel cell current density increased when the applied cell voltage decreased. The rule has been proven for the operating cell voltage of 1.1 V and 0.7 V, but then a sudden drop of the current value was noticed due

to probable lack of the fuel. In order to analyse the effect of fuel starvation as well as inlet concentrations of hydrogen and carbon monoxide further numerical studies need to be performed to estimate the impact of different feed compositions on SOFC performance. CFD results showed that the channel design plays an important role in the uniformity of mass fraction, temperature and current density distributions. In the areas where fuel was available, the fuel cell was electrochemically highly active and produced expected current at the applied voltage. In addition, it was demonstrated that the risk of hot spots decreased when a uniform distribution of mass fractions was assured. Therefore, providing a uniform distribution of the fuel cell operating parameters allows to reduce the risk of hot spots inside the fuel cell and as a consequence the fuel cell lifetime can be extended along with an increase of total power efficiency of the SOFC.

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SYMBOLS

F	Faraday's constant, C/mol
i	current density, A/m ²
i_0	effective exchange current density, A/m ²
n	number of electrons
R	universal gas constant, J/(kmolK)
T	absolute temperature, K

Greek symbols

α_{anode}	anodic transfer coefficient
$\alpha_{cathode}$	cathodic transfer coefficient
η_{act}	activation over-potential for H ₂ and CO oxidation reaction or O ₂ reduction reaction, V
ϕ_{cell}	electrical potential, V

Subscripts

an	anode
cat	cathode
CO	carbon monoxide
H_2	hydrogen
O_2	oxygen

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