CFD evaluation of solid oxide fuel cell performances with different fuels

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Abstract. The operation of solid oxide fuel cells (SOFC) fueling different types of carbon-based fuels (i.e. natural gas, biosyngas, coal gas, etc.) become one of the main topics of SOFC research in recent years. Fuel gases like methane can be steam-reformed or partially oxidized in the SOFCs. Hydrogen is produced from the reforming reaction, which has crucial impact on the performance of solid oxide fuel cells, especially on the current density, temperature distribution and the consequent thermal-stress. Mathematical modeling together with simulation is a useful approach for designing SOFCs and predicting their operation safety. With a single channel model taking direct internal reforming into account, this work aims to investigate the performance of an anode-supported intermediate temperature planar SOFC designed for methane under co-flow operation. SOFC performances with three different fuel gases are compared. The direct internal reforming (DIR) reaction of methane, the water-gas shift (WGS) reaction and the electrochemical reaction of hydrogen are the three reactions taken into account in this simulation work. Detailed comparisons of the gas concentrations and temperature change profiles among these three simulation models are presented and discussed. Thermo-dynamic predictions of both nickel oxidation and carbon deposition are also employed to check the operation safety of SOFCs fueled with different gases.

Keywords: SOFC, CFD, biosyngas

1. Introduction

SOFCs technology has been developed rapidly and is now commercially exploitable[1,2,3,4,5,6,7,8,9]. Electricity and heat can be cogenerated in SOFCs which are expected to be components of advanced energy systems because of its high efficiency, low pollution, and simplicity [1,2,5,6,9]. Significant research efforts have been made to exploit and develop the unique features of the SOFCs to obtain high efficiency compared with the conventional energy conversion technologies[4,6,7,8].

With internal steam reforming, SOFC systems own the potential to become a competitive technology. One of the essential aspects for the efficient operation of SOFCs is the suitable choice of fuel, and it is also becoming apparent that SOFCs are devices which can oxidize different types of fuel when they are sufficiently clean[10,11]. Biosyngas which is sustainable and practically CO_2 neutral is such a fuel. Therefore, biosyngas might act as an important fuel in the future. In the conventional SOFC anodes, the heat needed to sustain the endothermic reforming reactions is supplied by the exothermic electrochemical hydrogen oxidation and the water gas shift reaction[11,12]. In general, SOFCs are designed for methane and natural gases, and the SOFC performance with other fuels are still unknown and of much interests. When fuelling biosyngas into the SOFCs, whether they perform safely and efficiently as they do with methane and natural gase is an interesting question and need to be answered.

Mathematical models can provide a preview of cell potential, current density, different species concentrations and temperature, temperature gradient as functions of position, for various cell configurations and operating conditions, and be used to test the effects of changes on one or more variables and the relative system sensitivity to relevant design parameters[13,14,15]. The model can also be useful in predicting the possibilities of both carbon deposition and nickel oxidation, thus giving suggestions for safe fuel cell

operation. The main purposes of this paper are to: (1) present a steady-state model of a planar anode supported intermediate temperature SOFC designed for methane with DIR reaction by evaluating the representative single channel model; (2) compare the cell performances with different fuels; and (3) predict the possibilities of carbon deposition and nickel oxide formation which can damage the cell.

2. Model description

An SOFC designed for methane developed by ECN together with Delft University of Technology is employed for the calculations. For details, please see Qu et al[16]. A representative single channel unit that is assumed to be in the centre of a large stack, such that no end effects are present is considered in this work. Pressure drop along the channels is neglected. The geometric model represented in this work is shown in Fig.1. The present model is based on the co-flow design. The mathematical model is based on the conservation of mass, momentum, electrical charge and energy, coupled with appropriate constitutive laws. The electrochemical and steam-reforming models are specified in our previous work[16]. In this 3D CFD modelling, the solid and fluid domains are divided into discrete grid cells, as shown in Fig. 1(c). In each computational cell, the conservation equations of mass, momentum, species and energy are solved. The electrodes in the cell are treated as porous media. The additional source terms in the model are defined by our own user-defined functions. The extended model information can be found in the work of Qu et al[16].
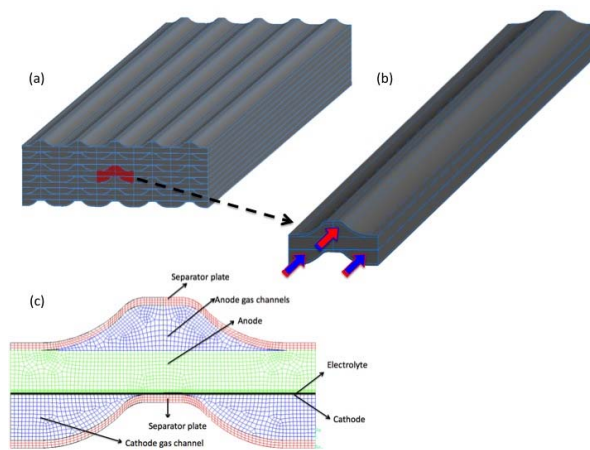

Fig.1. Corrugated single channel model: (a) Fuel cell stack schematic diagram; (b) single channel schematic diagram; (c) cross-section with grids for the single channel.

Some assumptions in this work: [1] only steam reforming and water gas shift reactions are being considered associate with hydrocarbon; [2] hydrogen is the only electrochemically oxidized fuel in this model, all CO and methane is firstly converted to hydrogen and then release electrons; [3] water gas shift reaction reaches the equilibrium rapidly and is always considered as at equilibrium[16]. The considered reactions can be found elsewhere [16].

3. Gas species concentrations

The SOFC is designed for methane originally, and the cell performances fuelling three different kinds of gas are compared in the model to see whether it works as efficiently as it does with methane. The gas compositions of three different fuel gases are specified as below in Table I[17,18,19].

Tabel I. Gas compositions [%]	CH_4	H_2	CO	CO_2	H_2O	N_2
Biosyngas	2	18	18	13	10	39
Methane	33	1	---	---	66	---
Pre-reformed methane	27.95	12	0.59	0.80	58.66	---

4. Cell performance

The Navier-Stokes and transport equations are solved with the finite volume method by using a commercial computational fluid dynamics simulation code, FLUENT, coupled with the user-defined electrochemical and the steam reforming reaction model to investigate the operating parameters of the proposed SOFC. The remaining physical properties (gas stream densities, heat capacities, and thermal conductivities) and gas velocities are determined for the inlet conditions, assuming ideal gas behavior, and are taken as constant values that apply throughout the system. The effective diffusivity coefficients are calculated under the assumption of 30% ($\varepsilon = 0.30$) electrodes porosity. All the plots presented are for the reaction zone (interface of the anode and the electrolyte). The fuel and oxygen gases are under co-flow. This section is divided into two sub-sections: Section 4.1 presents the simulated curves for gas species concentration as a function of the distance; Section 4.2 analyses the temperature gradients of a co-flow SOFC system with different fuels.
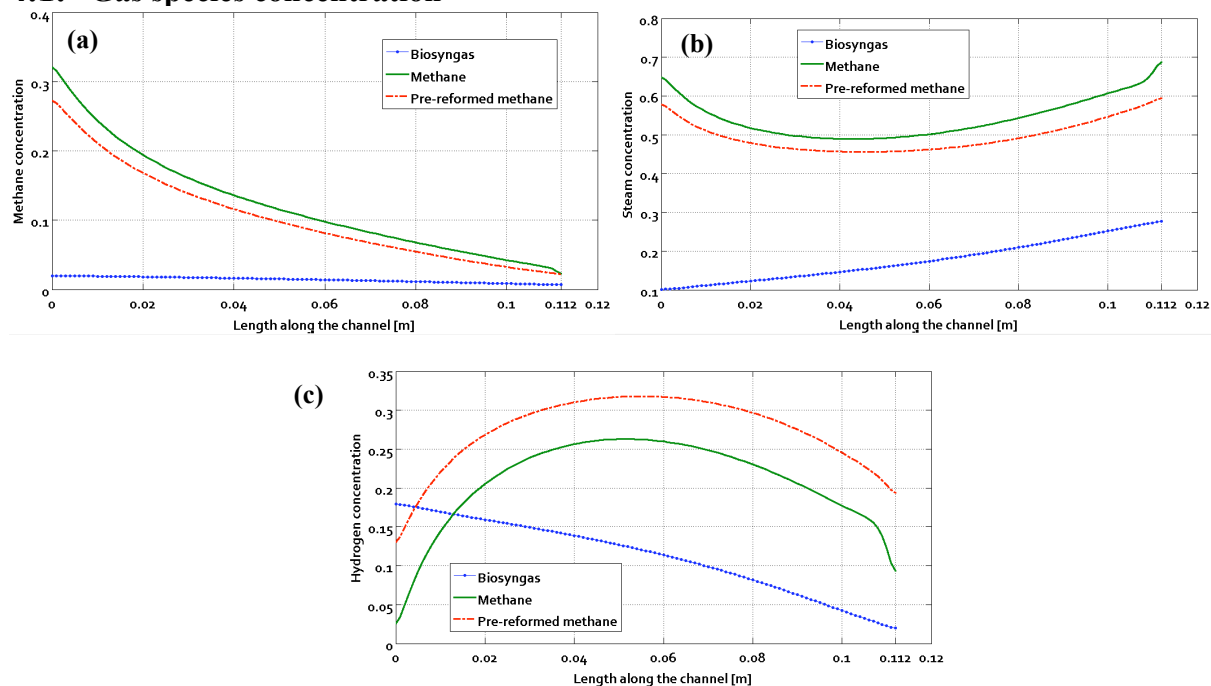
4.1. Gas species concentration

Fig. 2. Comparison of the gas species concentrations along the length of the channel for the three cases: (a) methane; (b) steam; (c) hydrogen.

The composition changes of methane, hydrogen and steam in three cases are illustrated in Fig. 2. The methane concentrations in all three cases only decrease due to the steam reforming reaction shown in Fig 2 (a). As shown in Fig.2 (b), steam concentration only increases in biosyngas, but decrease first and then increase in methane-based gases due to the combination of steam reforming reaction and electrochemical reaction. This also explains the changes in hydrogen, shown in Fig 2 (c). The reasons for the differences are: (i) the different initial mole fractions of methane make the reforming reaction rates different; (ii) the different temperature caused by the endothermic steam reforming reaction and exothermic water gas shift and electrochemical reactions make the reaction rates different.

4.2. Temperature gradient

The comparison of temperature gradients along the channel from the inlet to the outlet of the three cases is shown in Fig. 3. Due to the endothermic steam reforming reaction, more attention should be paid to the entrance of the cell for the risk of the thermal stress induced crack of the cell for all cases in the anode as well as the electrolyte. The exit part needs more attention when fueling methane compared with that of biosyngas and pre-reformed methane. As for interconnects, which are made from metal with a good thermal conductivity, heat can be well dissipated through it, thus thermal stress is less important. On the cathode side, the average temperature of cathode gas lags behind that of the solid parts and anode gas. As air is functioning

as coolant, the temperature of the air channel is lower than the other positions. So less attention is needed for the cathode part compared to the anode part.
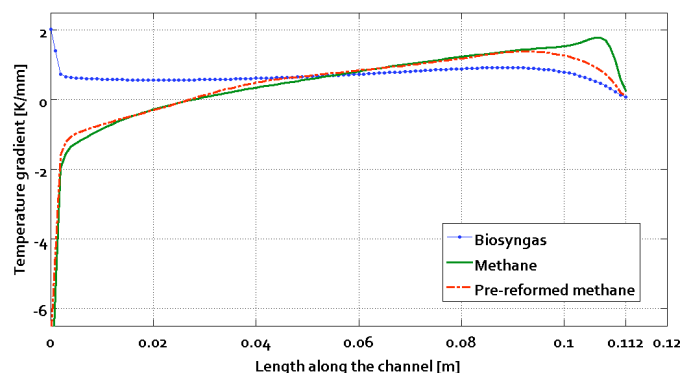

Fig. 3. Comparisons of the temperature gradients along the length of channel of the three cases.

5. Thermo-dynamic predictions

It is important to predict the possibility of the formation of NiO in anode materials, since it is one of the crucial signals of the degradation of fuel cell performance. In case of this research, gas concentrations in the exhausted fuel were used to investigate the degree of oxidation of Ni from existing oxygen in anode. Oxygen exists in fuel gas channel, and diffuses to the porous anode materials, which is made of Ni-based materials, and might have the chance to form NiO under certain conditions. However, due to the high steam-to-hydrogen ratio at the exit part in the anode gas channel, it is of first concern to clarify whether the commonly used Ni based anodes are stable in the metal phase with the increase of p_{O_2} from equilibrium with hydrogen and steam. The nickel oxidation thermo-dynamic prediction is shown in Fig. 4(a).

It is also important to predict the possibility of carbon deposition along the fuel channel. The deposition of the elemental carbon on the catalyst is a major issue for both the industrial steam reforming and the SOFCs with internal reforming. The carbon may be formed by the methane cracking reactions, or the Boudouard reaction. It is especially important to be aware of carbon poisoning when using Ni-YSZ, which is vulnerable to the carbon deposition. The thermo-dynamic prediction of carbon deposition is shown in Fig. 4(b).
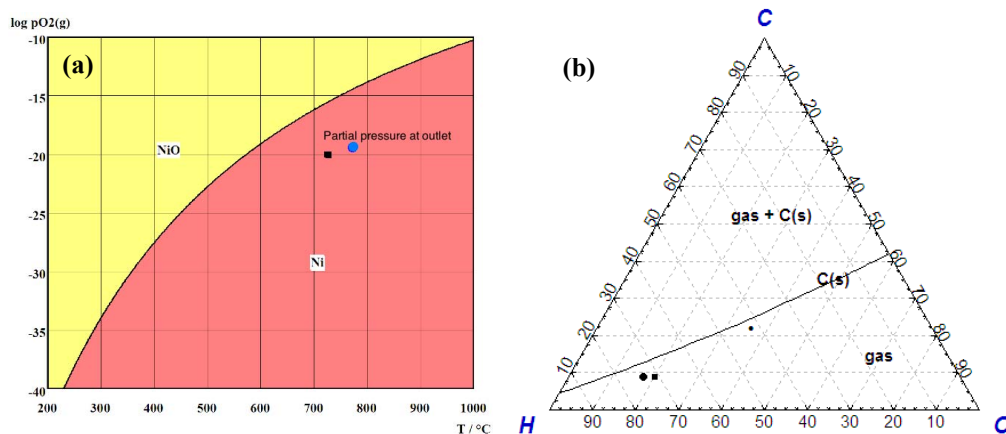

Fig. 4(a). Combined phase equilibrium diagram of Ni and NiO system from 200 ℃ to 1000 ℃ across an oxygen partial pressure ranges from 10^{-40} to 10^{-10} bar. "■" and "●" represent the highest oxygen partial pressure in the methane-based gases and biosyngas, under the given working condition; Fig. 4(b). Ternary diagram for operation with biosyngas, the solid line represents the deposition equilibrium boundary of carbon, above which is the solid phase region and below which is the gas phase region. The points "●""■""●"are the calculated positions according to the element fractions at the inlet for biosyngas, methane and pre-reformed methane.

Here in this work, anode materials stay in the metallic phase for all gases under the given condition as shown in Fig. 4(a).

Fig. 4(b) shows the predicted possibility of carbon deposition at a selected point in the cell; the solid line represents the carbon-deposition-boundary line. The possibilities depend on many factors, e.g., gas compositions and temperature distributions. The carbon deposition possibilities are calculated at the inlet where the chances for carbon deposition are higher along the channel. SOFCs directly fueled with three gases appear to be safe (under the boundary line).

6. Conclusions

The results in this work reveal that the SOFC designed for methane can work safely with biosyngas as well, under the same working condition. In summary, this work can draw conclusions as follows:

1. Different inlet gas compositions and concentrations lead to different reaction rates and different equilibriums.
2. Significant temperature gradients can be caused by either the combination or the interaction of the endothermic methane steam reforming reaction and the exothermic electrochemical reaction. Less attention is needed when fuelling biosyngas into the SOFCs.
3. Predictions of nickel oxidation and carbon deposition confirm the safety possibility of biosyngas fuelled SOFC with an SOFC designed for methane.

7. References

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