SIMULATION RESULTS AND EXPERIMENTAL VALIDATION OF SOFC OPERATION UNDER HYDROGEN

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ABSTRACT

An electrochemical model and a thermal model have been developed and coupled in order to study solid oxide fuel cell operation conditions depending on the nature of the fuel (4). A sensibility study performed on the model parameters has highlighted a significant contribution of the heat exchange coefficients. In order to validate these physical parameters, experiments have been performed at 800°C on anode supported cells upon hydrogen feeding. In the present work, special attention has been given to the measurements of the temperatures in the gas channel at the anode and cathode side. The elevation of these temperatures is found to be well correlated with the current density in the cell. Experimental temperature profiles along the gas channels are introduced in the thermal model of the cell as boundary conditions. Heat fluxes exhausted from the cell to the cathode and anode gas channel are simulated. These fluxes are compared to the heats evacuated by the fuel and air flows. This comparison is discussed according to the heat exchange parameter values.

INTRODUCTION

Upon solid oxide fuel cell (SOFC) operation, flow, thermal, chemical, and electrochemical systems are intrinsically coupled. Indeed, local heat generation and absorption affect the temperature distribution across the cell. Mathematical models allow understanding of the thermal and electrochemical behaviours of the cell which are essential for the SOFC developments. Some recent models have been developed in order to study the combined heat and mass transports in a planar single cell (1, 2). However, only little information is available regarding the validation these physical models (3). To study SOFC operation conditions, depending on the nature of the fuel, a specific model has been developed (4). In this paper, special attention has been paid to the validation of the thermal parameters of this model with experimental tests. In a first step, the validation has been achieved with cell operated under pure hydrogen.

EXPERIMENTAL

Three similar planar anode supported cells, manufactured by InDEC (cell type ASC1), have been tested. The anodic substrate is a sintered double layer made of a porous nickel cermet (Ni-8% mol Y_2O_3-doped ZrO_2) with a thickness ranging between 500 and 600 μm. The electrolyte is a dense YSZ layer 3 to 10 μm thick and the cathode layer is made...
of strontium doped lanthanum manganite (LSM) and has a thickness ranging between 30 to 50 μm. The cell diameter is 119 mm.

Cell tests have been carried out by flowing humidified hydrogen (3% H₂O) at the anode side at a flow rate of 6.10⁻⁴ mol/s (800 ml/min at room temperature). Dry air was sent to the cathode side at a flow rate equal to 1.5.10⁻³ mol/s (2000 ml/min at room temperature).

A schematic of the SOFC test system is given on Fig.1. Anode and cathode are fed at their periphery. Gas species react in the electrodes and are then exhausted at the centre. Four thermocouples are placed in the device to measure the temperatures of anode and cathode gases at both the inlet and the outlet gas channels (T₁, T₂, T₃, T₄). The cell is sealed in a ceramic holding ring. Gold meshes are used as current collectors. In order to improve the contact resistance between electrodes and current collectors, a compressive stress of 1160 Pa was applied on the gold meshes. However, we chose not to use any contacting paste in order to avoid any possible modification of the studied electrochemical properties of electrodes materials as recommended in (5). Finally, the cell tests were performed in a furnace at a controlled temperature of T_furnace=800°C.

![Figure 1. Test device used in the electrochemical measurements. Gas channels are represented. Inlet and Outlet gas temperatures are measured by four thermocouples (Tᵢ).](image)

**MODEL DESCRIPTION**

The model is composed of two combined parts. One describes the electrochemical behaviour of the cell, and the other its thermal behaviour. The circular geometry of the cell has allowed using an axisymmetric analysis (Fig. 2). A detailed description of the complete simulation model is given in reference (4).

The electrochemical model takes into account mass transport along the gas channel and diffusion through the porous and thick anode.
The electro-oxidation of hydrogen [1] is supposed to take place in the vicinity of the interface between the anode and the electrolyte:

\[
H_2 + \frac{1}{2} O_2 \leftrightarrow H_2O \quad \Delta H^\circ(1073)=-242.1 \text{ kJ/mol} \quad [1]
\]

The cell voltage, \( U_{\text{cell}} \), which is assumed to be constant along the radius, is imposed. The current density, \( i \), and the overpotentials, \( \eta_{\text{act}} \) and \( \eta_{\text{conc}} \), are calculated according to equation [2] by successive iterations:

\[
U_{\text{cell}} = E_{j=0} - (R_c + R_e)i - (\eta_{\text{conc,a}} + \eta_{\text{act,a}} + \eta_{\text{conc,c}} + \eta_{\text{act,c}}) \quad [2]
\]

With: \( E_{j=0} \) is the open circuit voltage, \( R_c \) the contact resistance between current collectors and electrodes, and \( R_e \) the electrolyte resistances.

\( R_e \) has been calculated, whereas all the other parameters have been fitted with experimental data. Their values are given in Table I.

**Table I. Electrochemical parameters.**

| Parameter                          | Value  |
|------------------------------------|--------|
| Anodic exchange current (mA \( \text{cm}^{-2} \)) | 1000   |
| Cathodic exchange current (mA \( \text{cm}^{-2} \)) | 80     |
| Cathodic limiting current (mA \( \text{cm}^{-2} \)) | \( 10^4 \) |
| Contact resistance (\( \Omega \text{ cm}^2 \))    | 0.8    |
| Electrolyte resistance (\( \Omega \text{ cm}^2 \)) | 0.05   |

The thermal part of the model takes into account heat transport by:

- Conduction within (i) the porous electrodes, (ii) the dense electrolyte and (iii) the ceramic holding ring, with the associated heat conductivities \( \lambda_{\text{eff,an}} \), \( \lambda_{\text{eff,cat}} \), \( \lambda_{\text{electrolyte}} \), and \( \lambda_{\text{ring}} \),
- Natural convection between the ceramic ring and the atmosphere of the furnace with the associated heat transfer coefficient \( h_{\text{ring}} \).
- Convection between the electrodes and the gas channels. The associated heat transfer coefficients, \( h_{\text{anode}} \) and \( h_{\text{cathode}} \), are considered to be constant along the gas channels.

Inlet and outlet anode and cathode temperatures are taken as boundary conditions. The heat produced by hydrogen electro-oxidation is assumed to be located in the vicinity of anode/electrolyte interface. All the thermal parameters of the model are given in Table II.

|                  | \( \lambda_{\text{eff}} \) (W m\(^{-1}\) K\(^{-1}\)) | \( h \) (W m\(^{-2}\) K\(^{-1}\)) |
|------------------|-------------------------------------------------|-----------------------------------|
| Anode            | 3                                               | studied parameter                 |
| Electrolyte (6)  | 3.8                                             | -                                 |
| Cathode          | 1.8                                             | studied parameter                 |
| Ceramic ring     | 3.8                                             | 10                                |

The electrochemical part of the model has been achieved using MATLAB whereas thermal simulations have been performed with the software CAST3M based on the finite element method.

RESULTS

The experimental performances obtained under hydrogen with the three tested cells are presented in Fig. 3a. An acceptable reproducibility is obtained between the three but the area specific resistances (ASR) are found to range between 1.1 to 1.7 \( \Omega \cdot \text{cm}^2 \), values which are very high. One experimental polarisation curve is compared to a simulated one on Fig. 3b. To obtain a good agreement between the two curves, a value of the contact resistance between current collectors and electrodes, \( R_c \), equal to 0.8 \( \Omega \cdot \text{cm}^2 \) is necessary. It is sixteen times higher than the electrolyte resistance and represents the most important contribution of the area specific resistance of the cell. The contact resistance is a parameter which can vary over a large range, depending in particular strongly on the mechanical loading of the cell. Therefore, this result can be attributed to the too slight loading applied on the current collectors and by the fact that it had been chosen to avoid contacting pastes. Nevertheless, these measurements can be used for the targeted thermal validations.

The temperature evolutions, recorded with the four thermocouples located in the gas channels, are presented on Fig 4. They have been measured as a function of time at increasing current densities. The temperatures in the gas channels are observed to increase strongly when the current density is raised. This temperature elevation can be attributed to the heat released in the cell by the exothermic electrochemical reaction.

Measured temperatures in gas channels are compared to the values of the temperature calculated inside the cell at various current densities on Fig 4b. In this case, measured temperatures \( \Delta T_1, 2, 3 \) and \( 4 \) are introduced as boundary conditions in the model. As expected, this simulated temperature is found to be higher than the gas temperature in the channels. Indeed, the gas species in the anode and cathode compartments are warmed by
the heat dissipated by the cell. The temperature difference appears moreover to be reasonable and tends to validate the model.

Figure 3. Polarisation curves under hydrogen at 800°C. (a) Voltage and Power Density as a function of current density. The reproducibility is satisfactory; (b) Comparison between an experimental and a simulated polarisation curves, a good agreement is obtained for $R_c=0.8 \, \Omega \cdot \text{cm}^2$.

Figure 4. Temperature evolutions in the gas channel depending on the current densities; (a) $\Delta T$ measured by the four thermocouples as a function of time with steps at 0, 5, 10, 15 and 20 A; (b) Experimental temperatures measured at the inlet of the gas channels ($T_3$ and $T_4$) plotted as a function of current density and compared to the temperature calculated inside the middle of the cell with the model.

DATA ANALYSIS

Temperatures measured at the inlet and outlet gas channels have been converted into thermal powers dissipated in the gas and the corresponding simulated heat powers calculated.

**Experimental Thermal Powers**

Before being introduced in the cell, the temperature of air and wet hydrogen are increased from ambient to 800°C. Then, the inlet anode or cathode gas species pass along the
electrodes and their temperatures are increased up to outlet values (i.e. $T_3$ and $T_4$). The thermal powers, $Q_{\text{anode}}$ and $Q_{\text{cathode}}$, required to induce these temperature elevations can be directly calculated using the following equations:

$$Q_{\text{anode}} = \dot{n}_{\text{H}_2/\text{H}_2\text{O}} \cdot C_p^{\text{H}_2/\text{H}_2\text{O}} (T_4 - T_{\text{furnace}})$$

$$Q_{\text{cathode}} = \dot{n}_{\text{air}} \cdot C_p^{\text{air}} (T_3 - T_{\text{furnace}})$$

with $\dot{n}_{\text{air}}$ and $\dot{n}_{\text{H}_2/\text{H}_2\text{O}}$ the molar flow rates (mol/s) of air and fuel, $C_p^{\text{air}}$ and $C_p^{\text{H}_2/\text{H}_2\text{O}}$ the molar heat capacities (J/mol.K) for air and H$_2$/H$_2$O mixture, $T_3$ and $T_4$ the experimental outlet temperatures.

Experimental thermal powers are plotted as a function of current density in Fig. 5:

![Figure 5. The anodic and cathodic “experimental thermal powers” represent the thermal fluxes which are required to heat the gas from 800°C to the measured temperatures at the outlet of the gas channels ($T_3$ and $T_4$).](image)

**Simulated Thermal Powers**

Taking the measured temperature as boundary conditions, the thermal field across the cell can be calculated using the model and the anode and cathode thermal powers dissipated from the cell to the channels can be extracted. In order to determine these thermal powers $Q_{\text{anode}}$ and $Q_{\text{cathode}}$, the following equations have been used for the finite element implementation:

$$Q_{\text{anode}} = \int_{\text{surf},a} \hspace{1cm} \left( T_{\text{gas channel},a} - T_{\text{surf},a} \right) \cdot h_a \cdot ds$$

$$Q_{\text{cathode}} = \int_{\text{surf},c} \hspace{1cm} \left( T_{\text{gas channel},c} - T_{\text{surf},c} \right) \cdot h_c \cdot ds$$

with $T_{\text{surf},a}$ and $T_{\text{surf},c}$ the anode and cathode calculated surface temperatures, $T_{\text{gas channel},a}$ and $T_{\text{gas channel},c}$ the temperatures of the anodic and cathodic gas channels linearly interpolated between the measured boundary temperatures.

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The values of these simulated thermal powers as a function of the heat transfer coefficients are presented on Fig. 6. In each case a set of \((h_a ; h_c)\) couples has been considered.

The anodic thermal power is found to decrease rapidly when the cathodic heat transfer coefficient is increased (Fig. 6a). Indeed, in this case, the heat is completely evacuated in the cathode channel. Similarly, high values of anodic heat transfer coefficients lead to the negligible thermal power at the cathode side (Fig. 6b).

**DISCUSSION**

Anode and cathode thermal powers have been evaluated i) directly from the experimental measurements and ii) thanks to the model, with the experimental measurements taken as boundary conditions. The best agreement between both approaches is obtained for values of \(h_{\text{anode}}\) approximately equal to \(h_{\text{cathode}}\), lower than 100 W/(m².K). As an illustration, a comparison between the experimental and simulated anodic thermal powers as a function of the current density is given on Fig. 7 for two specific values of \(h\). It is worth noting that the experimental and simulated curves exhibit a similar evolution. Furthermore, the values calculated either with the model or directly from the experimental data, have the same order of magnitude. However, the experimental thermal powers are always lower than simulated ones, even with heat transfer coefficients drastically decreased to 1 W/(m².K). This discrepancy could be explained by the fact that in its current state, the thermal model only considers thermal exchanges (i) by conduction with the ceramic holding ring and (ii) by convection with air and fuel. Several other types of heat exchanges can reasonably be expected to occur in the test system. Indeed, a fraction of the heat exhausted from the cell could be evacuated by conduction within the current collectors. In addition, radiative exchanges with the test system may not be negligible at 800°C. Consequently, the simulated thermal power, responsible for the obtained warming the anode and cathode gases, are overestimated.
CONCLUSIONS

A combined electrochemical and thermal model has been developed to study SOFC operations (4). In this work, experimental tests have been performed in order to validate the thermal part of the model under hydrogen operation. Local temperatures have been measured in the anode and cathode compartments during cell tests. These measurements have allowed determining experimental thermal powers dissipated from the cell to the gases. These values have been compared to thermal powers extracted from the model. The globally good agreement between the orders of magnitudes and the current densities dependence is obtained. The slight difference between the values can be explained by the assumptions of the model. A better prediction should be obtained by adding some improvements to the model such as the radiative and conduction heat exchanges with the test system surrounding the cell.

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