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Numerical Simulation of Reactive Transport Phenomena in the Hexis SOFC Stack

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An advanced computational reactive flow code has been implemented into Mathematica® to analyze (electro-) chemical conversions and various transport phenomena in the Hexis® SOFC-stack. The finite element approximation is applied for the space discretization of the continuity equations. The convection dominant cases are stabilized by the SUPG (Streamline Upwind Petrov Galerkin) method. The fluid flow in the interconnect channels is derived analytically with accurate representation of the slip velocity on the porous electrode surfaces. The mathematical system is linearized by means of a pseudo-time step discretization via a semi-implicit scheme. This stable iterative procedure is applied to an evolutionary problem and it converges unconditionally to the steady solution. Due to the low aspect ratio of the thickness to the cross-section of the cell, a one-dimensional model of the cell is coupled with the flow inside the gas channels. This approach reduces the computing time by retaining the main geometric details.

Introduction

Due to its novelty, there are still open questions about the behavior of SOFC-systems and numerical simulations appear as a valuable tool to predict their performance. A challenging task in this way is the development of reliable and stable numerical methods to explore the different geometric design features, to account for the electro-thermo-fluidic coupled fields and to have a realistic insight into the multi-component reactive-transport. Especially under extreme conditions where for example the fuel gets depleted, classical numerical schemes such as the Newton-Raphson method become unreliable. This is because these methods are not unconditionally stable.

In this study an advanced computational reactive flow code has been implemented into Mathematica® to analyze (electro-) chemical conversions and various reactive transport phenomena in the Hexis SOFC-stack. The physical model is governed by a system of nonlinear partial differential equations describing the conservation laws for mass, charge, energy and species. Multi-component diffusion fluxes are described by the Stefan-Maxwell equations. The fluid flow in the porous electrodes is described by the Darcy law.

To reduce the computing time without losing consistency certain geometric restrictions are assumed. For example, a low aspect ratio exists between the thickness and the cross-section of the cell. Therefore, the cell is treated in one dimension only and is coupled with the flow inside the gas channels.

This paper is organized as follows: In the first part we introduce some general hypothesis of the model including the geometric features, the transport model in the stack and an analytical model for the gas flow in the channels and the porous electrodes. This model includes an accurate description of the slip velocity at the porous wall. The coupling concept between channel flow and the transport in the cell is presented in the second part. In the third part we describe briefly the numerical methods that we applied in the Mathematica implementation. We introduce in the 4th part some selected results. Finally some conclusions and outlook perspective are cited.

The Model

We introduce an overall describing of the transport modeling in the cell and the channel with some natural assumptions based on the geometric feature of the stack design.

The Geometry

The design of the Hexis-SOFC is based on the concept of Planar Electrolyte Supported Cells; the full stack has a cylindrical shape with about 10 cm of radius and about 30 cm of height. In Figure 1, an illustration of the SOFC process is presented. In order to have an insight into the various fields involved in the SOFC process, assuming axial symmetry in the stack design, we adopt a simplified two-dimensional (2D) domain representing the vertical cross section through the center (Figure 2).

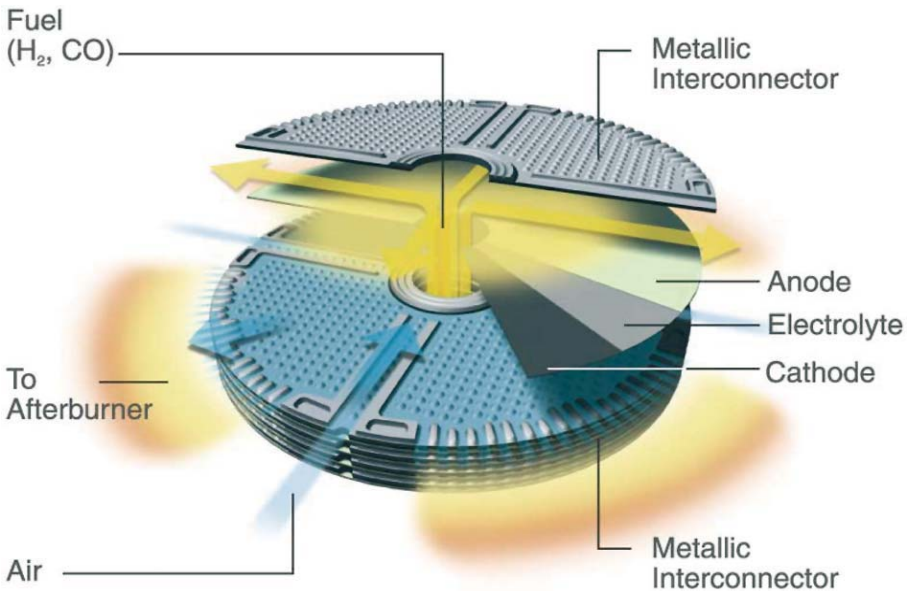

Figure 1. A description of SOFC process (Courtesy Hexis AG).

The dimensions of the different parts of the cell are introduced in Figure 2. A low aspect ratio is observed between the cell thickness and the channel length. Therefore, variations of all fields in the thickness direction of the cell (vertical direction) will be considerably larger than in the horizontal direction. This justifies the choice of the one dimensional (1D) approach of the transport in the cell thickness direction and subsequently allows an important reduction of the computational costs.

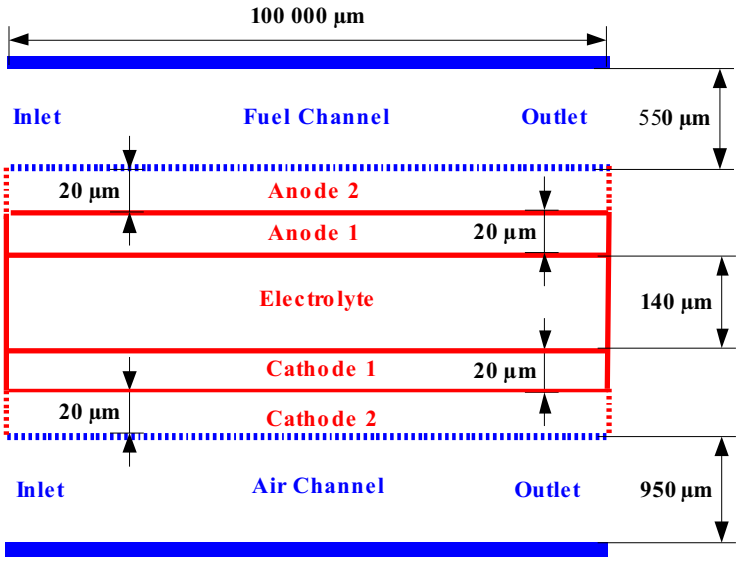

Figure 2. A schematic representation of the parts of SOFC taken in vertical cross section.

Transport Phenomena inside the Stack

Briefly, the gas and charge transport in the cell is based on the following assumptions: steady state flows, conservation of total mass and charge. Gas transport by convection, bulk diffusion and Knudsen diffusion (surface diffusion is neglected). For multi-species bulk diffusion flux the Stefan Maxwell equations are used. The Darcy flow in the cell is assumed to be incompressible, whereas the open flow in the channel is considered to be compressible.

Analytical Model for Compressible Channel Flow

The gas flow in the channels with porous walls has been studied by several authors, see e.g. (2). An important feature of such flow is the non-zero velocity at the porous boundaries which is called slip velocity. Roughly speaking, a slip boundary condition appears on the surface which constitutes the contour of a porous body, through which the fluid flow take place also. In several known approaches, the slip velocity is taken as a *a priori* known quantity and it usually serves as a boundary condition to solve the momentum conservation equation like Navier-Stokes. The limitation of those approaches is that slip conditions are most frequently estimated in an empirical way and do not necessarily correspond to all real situations.

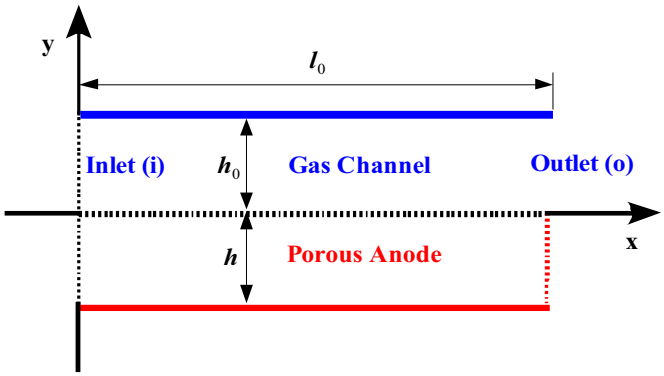

Figure 3. A channel flow with a permeable wall over a porous anode.

The flow model applied in this work is based on the Djordjevic's approach (1). Compressible flow in a channel with one permeable wall is successfully treated as the problem of strong interaction between the fluid flow in the channel region and the fluid flow in the porous region. A simultaneous solution of the equations describing the flow in both media are carried out. At the interface the slip velocity is considered as an arbitrary quantity, not known a priory and which can be determined by matching the shear stresses on the porous wall of the channel. In this way, the need for using the empirically defined boundary conditions as those of Beavers and Joseph (3) is eliminated. Our aim here is to present the final results of the analytical approach, for more details reader can see (1). Thus we need to introduce the following notations (Figure 3):

- l_0, h_0 and h: the channel length, the channel height and the electrode height,
- p_i, p_o and u_o: the inlet pressure, the outlet pressure and the outlet average velocity,
- μ and $\tilde{\mu}$: the fluid viscosity and the effective viscosity in the porous electrode,
- κ and $\tilde{\kappa} = \kappa\,\tilde{\mu}/\mu$: the permeability and the relative permeability of the porous electrode.

We suppose isothermal conditions for the transport processes. Regarding the low aspect ratio between the channel height and the channel length, we consider the convective mass flux only in the longitudinal direction. According to (1), the pressure p is approximated analytically as:

$$\frac{p^2}{p_o^2} = P^2 - \left(P^2 - 1\right)\frac{x}{l_0}, \quad \text{with} \quad P = \frac{p_i}{p_o} > 1. \qquad [1]$$

The slip velocity at the channel-electrode interface is obtained in the form:

$$u_{slip} = \frac{(P^2-1)\,p_o u_o}{4p}\,\frac{2\left(\cosh(\sigma H)-1\right)+\sigma\sinh(\sigma H)}{\sigma\cosh(\sigma H)\left(\tilde{\lambda}\,\sigma+\lambda\tanh(\sigma H)\right)}, \qquad [2]$$

where

$$H = \frac{h}{h_0},\ \sigma = \frac{h_0}{\sqrt{\tilde{\kappa}}},\ \lambda = \frac{\mu\, l_0 u_o}{p_o h_0^2} \text{ and } \tilde{\lambda} = \frac{\tilde{\mu}\, l_0 u_o}{p_o h_0^2}. \qquad [3]$$

In the channel, the velocity component in x direction is obtained as:

$$u = \frac{u_{slip}(h_0 - y)}{h_0} - \frac{y(h_0 - y)}{2\lambda h_0^2}\,\frac{p' l_0 u_o}{p_o}, \quad \text{with} \quad p' = \frac{\partial p}{\partial x}. \qquad [4]$$

In the porous electrode, the velocity component in x direction is written as:

$$u = \left(\frac{u_{slip}}{\tanh(\sigma H)} + \frac{\left(\cosh(\sigma H)-1\right)h_0^2 p'}{\sinh(\sigma H)\sigma^2\tilde{\mu}}\right)\sinh\left(\frac{\sigma y}{h_0}\right) + \left(u_{slip} + \frac{h_0^2 p'}{\sigma^2\tilde{\mu}}\right)\cosh\left(\frac{\sigma y}{h_0}\right) - \frac{h_0^2 p'}{\sigma^2\tilde{\mu}}. \qquad [5]$$

One easily observes that the velocity component u in x direction in both media takes the value of the slip velocity at the channel-electrode interface (where $y = 0$). On the upper channel wall (where $y = h_0$) and on the bottom of the electrode (where $y = -h$) the velocity u is zero. Considering the geometric features of the SOFC channel, it is shown analytically in (1) that the velocity component in y direction is negligible.

The Coupling Concept

In this paper we present a new, simple and practical way to account for the interactions between the "in cell" fields and the gas channel flow.

A Simple 1+1D Cell-Channel Model

It is natural to consider the 2D transport in the channel as convection dominant in the longitudinal direction and as diffusion dominant in the transverse direction (toward the cell). A common approach is to represent the in-channel transport phenomena by a one-dimensional (1D) convection diffusion model (Figure 4). In this perspective, we only consider the average value of the longitudinal component of the flow velocity. Furthermore, the species diffusion fluxes in the transversal direction (i. e. the species exchange with the cell) are applied as source/sink terms for the 1D Channel Model.

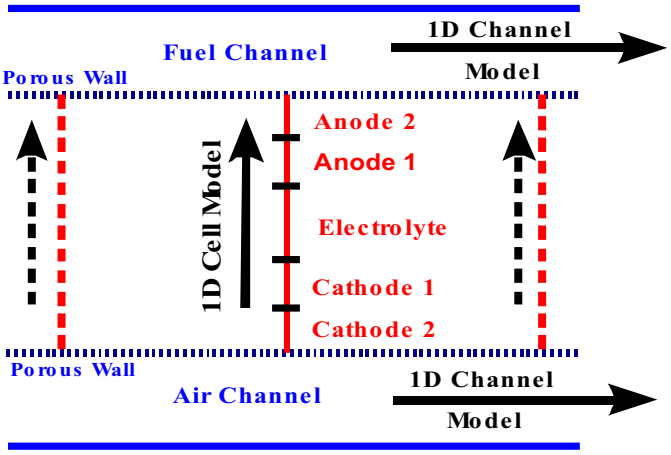

Figure 4. A simple 1+1D cell-channel modeling of a cell unit.

Therefore the 1D Cell and the 1D Channel transport problems are coupled in a simple 1+1D Cell-Channel Model, see Figure 4. The whole problem can then be solved by establishing an iterative computation between the 1D Cell and the 1D Channel Models.

The Boundary and Central Flow in the Channels

Since the species distribution along the channel-electrode interface play a decisive role on the magnitude of the diffusive flux in the cell, the simple 1D simulations in the channel, as introduced above, still have a limited representation of the process. For a yet simple, but more realistic approach, we investigate the species transport inside the gas channel at two levels (Figure 5): firstly we consider a 1D Channel transport problem located on the central line of the channel where the velocity field $u_{C.L.}$ is estimated by its average cross-sectional value i.e.

$$u_{\text{C.L.}}(x) = \frac{1}{h_0}\int_0^{h_0} u(x,y)\,dy \qquad [6]$$

and by applying as source/sink terms the outer flux resulting from the 1D Cell problem.

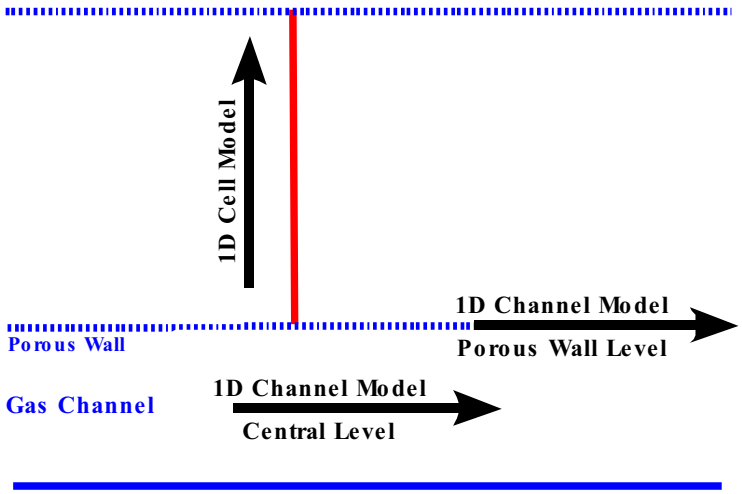

Figure 5. A cell-channel modeling, the 1D channel transport is solved in two levels.

Secondly, we consider a 1D Channel transport problem located along the porous wall of the channel where the species are driven by the slip velocity. In consistency with the continuity of the diffusive flux crossing the channel-electrode interface, we propose a zero source term for the 1D Channel problem on the porous wall.

Regarding the low aspect ratio between channel height (h_0 ~ 500 μm) and channel length (l_0 ~10^5 μm), the boundary values of the two mentioned 1D Channel problems are approximately related to the cell diffusive flux at the porous wall:

$$U_{\text{P.W.L.}} - U_{\text{C.L.}} \approx \frac{h_0}{2}[D]^{-1}.J\,, \qquad [7]$$

where $[D]$ is the diffusion tensor, $U_{\text{P.W.L}}$ and $U_{\text{C.L.}}$ are the species concentration at the porous wall level (P.W.L.) and the central level (C.L.) respectively and $\boldsymbol{J}$ is the cell flux located at the porous wall at the outlet of the channel.

The Cell-Channel Coupling

As presented in Figure 6, in each iteration step, we start by solving the 1D Channel problem on the center level. The boundary values are used to update those on the boundary of the channel-electrode interface via equation (1). Then we solve the 1D Channel problem at the porous wall level and the obtained value is used as Dirichlet boundary condition for the 1D Cell problem. In a new iterative step, we solve the cell problem and the outer species flux is applied as a source term for the 1D Channel on the central level. Note that in each step the species flows are computed in both the anodic and the cathodic gas channels.

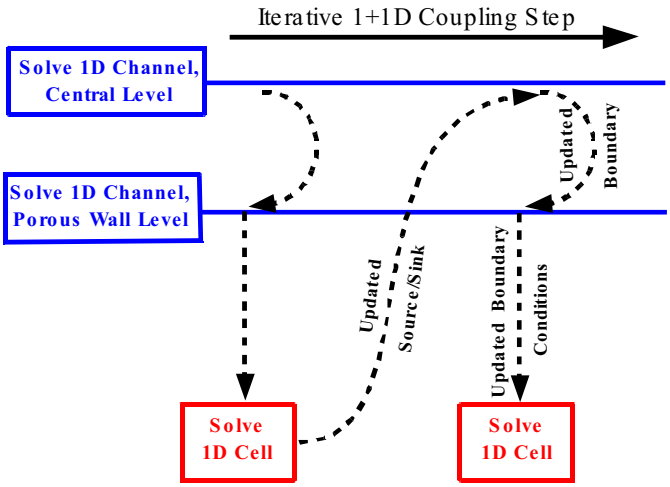

Figure 6. An iterative concept for solving 1+1D cell-channels model.

The New 1+1D Model Compared with the Existing 2+1D Model

The coupling between cell and channel was already studied within the framework of a 2+1D model where the channel is taken as a 2D planar domain perpendicularly to the 1D Cell direction (4). In this approach the effect of the channel height doesn't appear and the 2D flow in the channel is presented as an average with respect to the channel height. Although it is computationally efficient, the limitations of such a model are in the lost information about the interfacial distribution of the species on the porous wall of the channel (channel-electrodes interface). Especially the slip velocity in the channel porous wall is not described in (4).

The Numerical Methods

The Nonlinearity: Pseudo Time Step

The mathematical system is linearized by means of a pseudo-time step discretization via a semi-implicit scheme. This stable iterative procedure is applied to an evolutionary problem and it converges unconditionally to the steady solution. This is a significant advantage over other solving schemes for non-linear problems such as the Newton-Raphson method where the convergence depends on the initial conditions provided by the user.

The SUPG Stabilization for Convection Diffusion Transport

The finite element approximation is applied for the space discretization of the continuity equations. The convection dominant cases are stabilized by the SUPG (Streamline Upwind Petrov Galerkin) method (5). Since we are dealing with a 1D problem, the “numerical diffusion” resulting from SUPG method still is enough to avoid any oscillations in the results.

The GLS Stabilization of Darcy Problem

In the study of a simple 1D Cell Model uncoupled with the channel, Darcy flow in the electrodes is explored. To insure the conservation properties in the results when solving

for the Darcy flow, a stabilized mixed finite element method GLS is introduced in the implementation of the numerical methods. The flow problem is solved in the whole cell and a penalization of the velocity is applied in the impermeable subdomains.

The Simulations

The iterative computation of the channel-cell system takes about two minutes on a current, single-CPU PC (2.4 GHz). The convergence is reached after 7 global iterative coupling steps (see Figure 6) and after 10 pseudo time steps.

In Figure 7 we introduce the velocity field in the anodic channel, as well as the corresponding slip velocity at the porous wall. One sees that the slip velocity has a weak dependence on the location along the channel, caused by the compressibility effects, but strongly depends on the porosity of the adjacent electrode.

In Figures 8-10, the variations of various gas species concentrations are shown along the gas channels. The values denoted by the subscript "C.L." represent the vertical averages of the species concentrations in the channels. The subscript "P.W.L." denotes the values at the interface between a porous electrode and the open gas channel.

Note that in the displayed test case, boundary conditions at the central levels are provided for both the inlets and the outlets of the anodic and cathodic gas channels. Due to fuel consumption and the generation of oxidation products within the active layers of the cell, we observe a considerable difference between the species concentrations on the central and porous wall levels. The results shown in Figures 11 describe the variation of the ionic and electronic potentials along the SOFC.

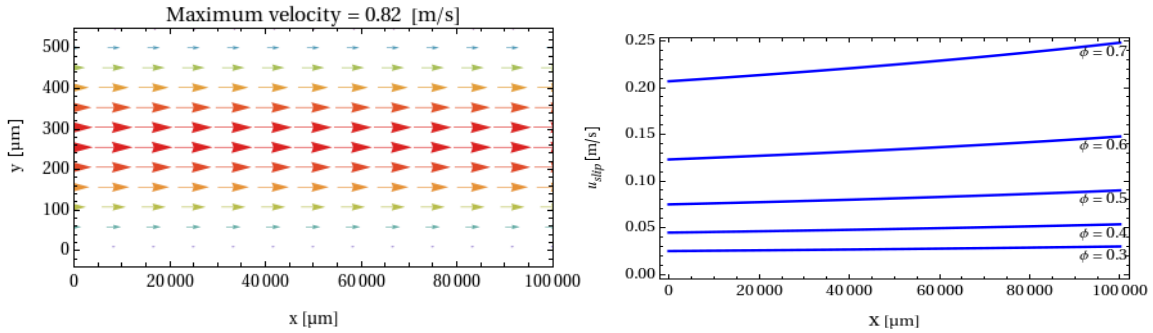

Figure 7. Gas velocity in the anodic channel (left). Slip velocity for different values of anode porosity ϕ (right).

In order to exhibit the effect of the channel geometry, we reproduce the iterative calculation under the same conditions with choosing 300 µm height for the channels. In Figure 12 we introduce the species distributions along the anodic channel. A comparison with Figure 8 shows significantly the impact of the channel geometry on the results.

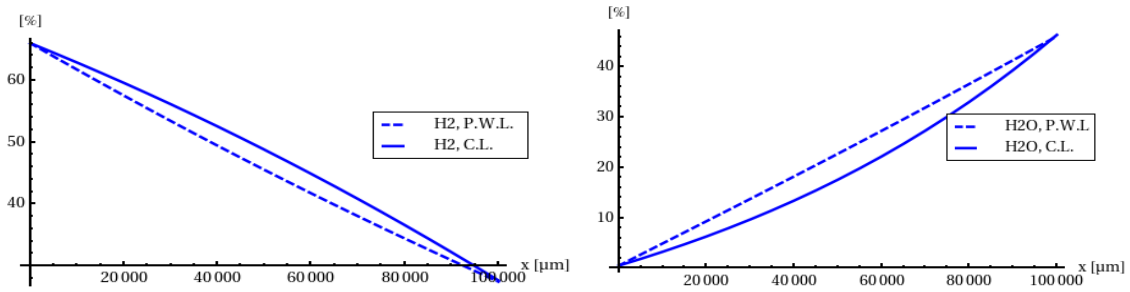

Figure 8. The concentration of Hydrogen (left) and Water (right) in the anodic channel on the porous wall (P.W.L.) and the central levels (C.L.), see Figure 4.

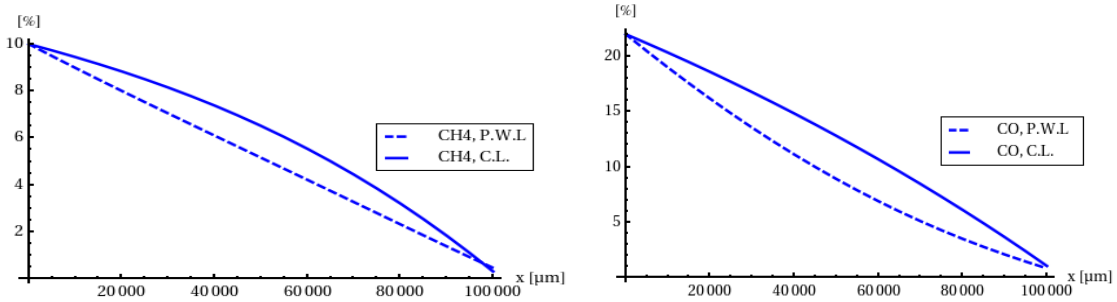

Figure 9. The concentration of Methane (left) and Carbon Monoxide (right) in the anodic channel on the porous wall (P.W.L.) and the central levels (C.L.).

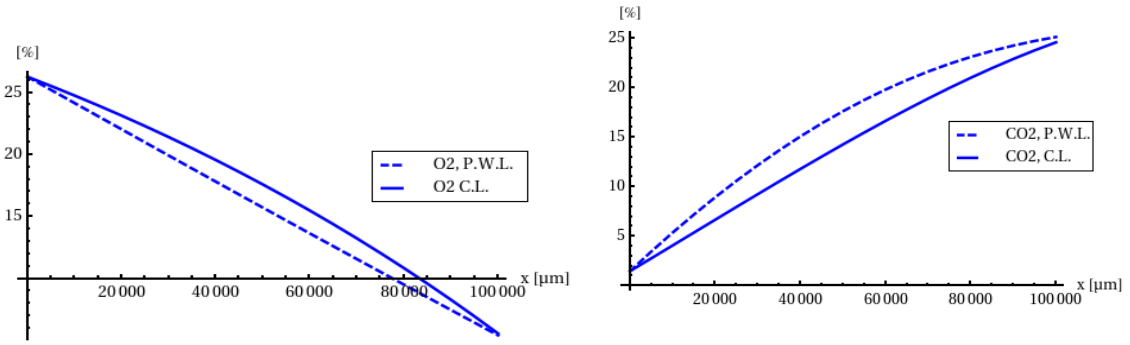

Figure 10. The concentration of Oxygen in the cathodic channel (left) and Carbon Dioxide (right) in the anodic channel.

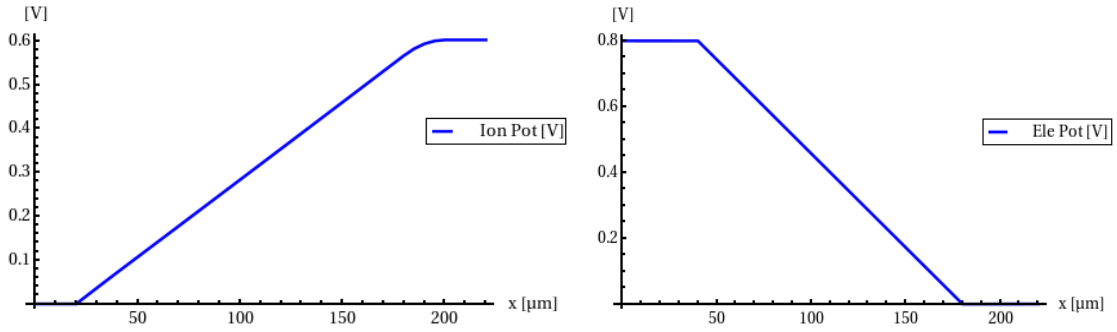

Figure 11. The ionic potential (left) and the electronic potential (right) in the cell.

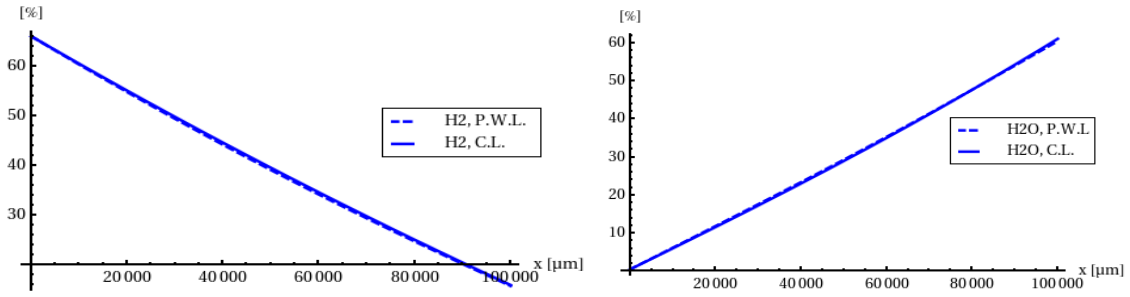

Figure 12. The concentration of Hydrogen (left) and Water (right) in the anodic channel with height = 300 µm.

Conclusions and Perspectives

In this paper we have shown the efficiency of the 1+1D model to study the reactive transport phenomena in the HEXIS-SOFC via a unconditionally stable pseudo-time scheme. The numerical technique have been implemented into Mathematica are suitable for the simulation of several repeat units. The computation time for the presented Cell-Channel Model is about 2 minutes on standard PC. This allows one to perform full stack simulations in a reasonable amount of time. The presented analytical formulation of the slip velocity on the porous wall of the gas channel depends on the porosity of the fuel cell electrodes. It may be also used as a boundary condition for numerically solving 3D Navier-Stokes flows in channels with porous walls.

Acknowledgments

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