MODELLING OF PLANAR SOFC

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ABSTRACT

SOFC modelling was performed on two different levels. Micromodelling, which is mainly concerned with electrochemical kinetics, potential field distribution and mass flow in the electrode structure taking into consideration the actual geometry of the PEN structure. This approach is important for investigations of electrochemical kinetics. Macromodelling aims at the operating conditions of the whole stack. In this way current and potential distributions are obtained in the cell and the stack. In addition, the local temperatures and the gas composition under various load conditions can be calculated. The results for the steady state and transient conditions are important informations with respect to the stack design.

Key words: micromodelling, stack modelling, SOFC, transient conditions

INTRODUCTION

There are two different fields for modelling of the electrochemical cells: micromodelling, which deals with the internal geometry of the single cell, especially the porous structure of the fuel cell electrodes taking the reaction kinetics into consideration. This kind of modelling can be used either as a basis for the macromodelling computations or as a tool for a more precise description of the usual electrochemical investigation techniques, e.g. impedance spectroscopy of the porous electrode systems. The second kind of modelling is macromodelling, which primarily deals with the engineering aspects of the electrochemical cell design. Both of them use the mathematical description of the heat, mass and charge distribution in the given geometric area of the SOFC system.
ELECTROCHEMICAL MICROMODELLING

The Simulation Program

The computation of the heat, mass and charge distribution in the elements of the single fuel cell is influenced by the material, geometric and processing data, as well as the characteristics of the actual electrochemical reaction. The starting point of the modelling involves the full description both of the cell geometry and the reaction kinetics. Using the fundamental laws of energy, mass and charge conservation, the temperature, concentration and potential distribution in the single cell unit are calculated by the finite integral technique (1). The resulting balance equation systems for the flows are solved by a multigrid algorithm (2). Adiabatic calculations are possible as well as calculations considering the heat exchange with the surroundings. Spatial periodic boundary conditions are assumed to account for the modular design of the stack. An average value of the heat capacity is used for the convective heat transport in the gas channels.

Computation in the Anodic Structure

The boundary condition for the potential simulation is a constant potential difference between the bipolar plates. This operating voltage is generated by the Nernst equation less the ohmic losses in the cell and the overpotentials at both electrodes. The local overpotential depends on the current density across the phase boundary and is given by the local potential difference between the electronic and ionic conductor. Additionally to this, the individual potentials of both the metal and the electrolyte are calculated for the anode in the same volume element. In each volume element a uniform distribution of the metal and electrolyte phase as well as a constant porosity are assumed.

The operating data used for the calculations are: gas inlet temperature 1173 K, mean porosity 40%, the exchange current density varies between 35 and 350 mA/cm². The mean conductivities of the electrolyte and the nickel phase in the bulk of the anode are 0.1 S/cm and 500 S/cm, respectively.

Fig. 1 and Fig. 2 show the current density distribution in the electrolyte phase of the anode (penetration depth of the electric field) along the gas flow direction and the thickness of the electrode. The co-flow cell operates at 1173 K air inlet temperature and 0.7 V output voltage. Fig 1 shows that at the high exchange current density \( j_0 \) of 350 mA/cm², the charge transfer between the Ni-phase and the YSZ-electrolyte phase only takes place near the phase boundary anode / electrolyte. This region has a thickness of 15 μm and the sum of the current density through the whole electrolyte phase is 45 mA/cm². If the exchange current density is reduced by a factor of 10 (Fig.2) the charge transfer takes place over the entire volume of the anode. Despite the fact that the
maximum current density in the electrolyte phase increases to a value of 75 mA/cm², the overall current density of the cell decreases due to the poorer electrode kinetics. Fig. 3 shows the comparison of the current density in the electrolyte phase of the anode with the overall current density at the phase boundary anode / electrolyte as a function of the exchange current density \( j_0 \). With decreasing \( j_0 \) the current density in the electrolyte phase of the anode increases. This shows the importance of the charge transfer in the anode volume in the case of unfavourable electrode kinetics.

**Cathode Kinetics**

For the correct calculation of the reaction kinetics, the knowledge of the possible reaction mechanism as well as its dependency on the temperature and on the corresponding gas partial pressures of the reaction gases is necessary. These parameter are used as the input data for the evaluation of the electrode kinetics in the SOFC cell by means of a suitable electrochemical technique, e.g. impedance spectroscopy.

In the case of oxygen reduction in the SOFC, different charge transfer mechanisms were proposed (3-5). As an example, the mechanism according to van Hassel et al. (5) was used. It is based on the following reaction steps. A dissociative adsorption of the molecular oxygen at the surface of the electrode pore is followed by the surface diffusion of the adsorbed oxygen towards the three-phase boundary cathode / electrolyte / gas, which is assumed to be one-dimensional. The next step is the electrochemical formation of the adsorbed oxygen ion \( O^- \) at the above phase boundary. This ion is finally reduced to the \( O^{2-} \) ion, which is mobile in the bulk electrolyte phase. Fig. 4 shows the corresponding surface degree of coverage by adsorbed \( O^- \) ion at the phase boundary cathode / electrolyte / gas. The coverage of the surface under the gas channels is nearly constant along the flow direction due to the low oxygen consumption in the fuel cell. Under the supporting current distributor, the degree of coverage decreases along the air flow. Across the width of the cell a minimum coverage appears at the edges of the channels. It can be shown that there are high \( O^- \) coverages in regions of low polarizations and vice versa. These calculations enable the correct evaluation of the electrochemical kinetics in the SOFC systems, as will be shown in a later contribution.

**STACK MODELLING**

**The Simulation Program**

In addition to the activities on modelling on the microscale the simulation of the whole stack is necessary to optimize the fuel cell. For this purpose a comprehensive computer program has been compiled (6,7) to simulate the operating conditions. All the field quantities, e.g. the molar flux distribution of the participating gas components, the current density, the gas and solid temperatures, and the Nernst potential, are determined.
in three dimensions, i.e. along and across the stack. Furthermore, transient processes can
be simulated since the equations of conservation of mass, energy and charge are
temporally resolved.

Three-dimensional Distribution of Temperature

Generally, a stack cannot be considered as an adiabatic system. Due to the high
temperature level the heat radiation from the stack surface to the surroundings may have
an essential effect on the heat balance of the system. This results from the fact that the
equivalent heat transfer coefficient for radiation is about $h = 470 \text{ W/m}^2 \text{ K}$ at 1000 °C.
Therefore a stack operating at such high temperatures transfers a large amount of heat
even at small temperature differences. This is demonstrated by the example of a
calculation performed for two 50-cell stacks, one consisting of ceramic, the other of
metallic bipolar plates. Both stacks are operated under the same conditions: the fuel -
30% pre-reformed methane - and the air enter the stack at 900 °C. The O$_2$ stoichiometry
factor is $\lambda = 7$, the fuel utilization $u_f = 0.85$ and the current density required $j = 300$
mA/cm$^2$. The stack is considered to be adiabatic except the top plate, which radiates
against a furnace temperature of 900 °C. Fig. 5a shows the result for the ceramic stack.
The temperature distribution is illustrated for a plane parallel to the fuel flow direction
and through the temperature maximum of the cell. It is evident that the cells in the
middle of the stack behave as adiabatic cells. The isotherms are parallel to the length
axis of the stack. A comparison of the temperature distribution in a horizontal central
cross section of the stack and for an adiabatic single cell indicate that they are identical.

Near the top and the end plates, however, the three-dimensional effect is visible
resulting from heat radiation at the top and heat conduction in the bottom plate. The
penetration depth of the effect is 5 to 7 cells. The temperature drop in the top cells is
associated with a decrease of the cell voltage. In the present calculation the top cell
voltage is by about 20 mV less than the mean value.

For the metallic stack the three-dimensional effect described above is more
pronounced (see Fig. 5b). Due to the high conductivity of the solid material the stack is
affected over the whole length by the heat removal from the top plate. About 25 % of
the waste heat is radiated to the surroundings which is higher by a factor of 4 than for
the ceramic stack. The temperature distribution along the stack, however, is more
uniform than for the ceramic stack.

Transient Behaviour During Load Change

For controlling purposes it is necessary to have information about the response of the
stack to load changes. Therefore calculations have been performed setting a step function
of the current density for the system. Fig. 6a shows the cell voltage for a stack where
the current density is instantaneously set to $j_2 = 500 \text{ mA/cm}^2$ starting from different
initial values $j_1 = 200, 300$ or $400 \text{ mA/cm}^2$. For $t < 0$ the cell voltage correlates with the
initial current density. At $t = 0$, where $j_2 = 500 \text{ mA/cm}^2$ is required, the voltage breaks down and shows an overshooting below the new steady-state cell voltage of $U = 0.59 \text{ V}$. The amount of overshooting increases with the magnitude of the current density jump. The relaxation time, however, is independent of that quantity.

The reason for the overshooting effect can be explained with a view to the temporal temperature development of the solid material exhibited in Fig. 6b. At $t = 0$ the cell is still at the initial temperature. The higher current density, however, causes a larger amount of waste heat which leads to a temperature increase associated with a feedback to the internal electrical cell resistance. Thus the variation of the cell voltage is closely related to the temperature of the solid structure.

In order to facilitate the prediction of the relaxation time an attempt was made to generalize the problem by introducing characteristic quantities. An analysis of the heat transferring processes yields two governing dimensionless groups. The first is the modified Fourier number, $F_o$

$$F_o = \frac{\Delta t \, a}{h_{\text{eff}}^2} \quad (1)$$

which contains the relaxation time, $\Delta t$, the thermal diffusivity, $a$, of the solid material and the effective thickness of the bipolar plate, $h_{\text{eff}}$, defined as

$$h_{\text{eff}} = (1 - \varepsilon) \, (d_b + h_a + h_f) \quad (2)$$

where

$$(1 - \varepsilon) = d_b + b_r (h_a + h_f) / (b_s + b_c) \, (d_b + h_a + h_f) \quad (3)$$

The second characteristic number is the Source Term number, $S_o$,

$$S_o = (1 - \eta) \, U \, j \, h_{\text{eff}} / (\eta \, \lambda_s \, \Delta T) \quad (4)$$

in which the waste heat rate, $U \, j \, (1 - \eta) / \eta$, the cell geometry, $h_{\text{eff}}$, the thermal conductivity, $\lambda_s$, of the solid material and the gas temperature increase, $\Delta T$, along the cell occur.

Computations have been conducted varying the geometrical and heat transfer conditions in a large range. The result plotted as $F_o$ vs. $S_o$ is shown in Fig. 7. It can be approximated by a simple relationship:

$$F_o = 0.72 \, S_o^{-1.1} \quad (5)$$

Eq. (5) enables the engineer to predict the relaxation time for arbitrary conditions of a planar SOFC without performing expensive computations.
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Fig. 1. Distribution of current density passing through the electrolyte phase in the anode at exchange current density $j_0 = 350 \text{ mA/cm}^2$

Fig. 2. Distribution of current density passing through the electrolyte phase in the anode at exchange current density $j_0 = 35 \text{ mA/cm}^2$
Fig. 3. Contribution of the current density through the electrolyte phase in the anode to the total current density as a function of exchange current density.

Fig. 4. Degree of coverage by the adsorbed $O^-$ ion at the three-phase boundary $cathode/electrolyte/gas$ across two air channels in the direction of gas flow.
Fig. 5. Temperature distribution along
a) ceramic stack
b) metallic stack

Fig. 6. a) Transient cell voltage during load change
Fig. 6. b) Temperature of solid during load change

Fig. 7. Fourier vs. Source Term number for estimation of relaxation time