Mathematical modeling aiming at SOFC design and operating condition optimization is conducted at the Forschungszentrum Jülich. Computational Fluid Dynamics (CFD) software is used for three dimensional (3D) stack modeling and a rapidly executing self-written one dimensional (1D) code for calculating "along-the-channel" profiles of e.g. the gas temperature of the fuel and the air, of the solid body, which is mainly composed of the bipolar (interconnector) plate, of the gas compositions (e.g. in molar fractions) and other interesting quantities. This co-simulation approach using both the 3D and the 1D tool was presented at the SOFC-VIII (1). This contribution discusses results regarding the non-isothermal Voltage-Current (V-I) behavior including dynamic load changes produced by using the 1D tool.

INTRODUCTION

V-I curves are usually measured in the laboratory using small single cell set-ups placed in a furnace and operated at low fuel utilizations $U_f$ (up to about 10%). In this set-up, the furnace ensures almost isothermal operating conditions while the V-I curve is usually measured at constant fuel and air flow. However commercially interesting stacks are not operated under these conditions: It must be possible to operate them self-sustained under approximately adiabatic (thermally insulated) conditions at technically interesting fuel utilizations of about 70 to 80%. Since a change of the electrical current output and the resulting change of the cell voltage alter both the electrical and the thermal energy release rate, the SOFC’s operating temperature will change, too. Hence isothermally measured V-I curves at low fuel utilization do not apply to these non-isothermally operated stacks any more. Since it is important to know the non-isothermal V-I behavior for proper systems design, it is interesting to gain some insight into non-isothermal SOFC behavior prior to carrying out expensive experimental work. The 1D “along-the-channel” model was used to run non-isothermal behavior simulations and the results will be discussed in this contribution.

Simulating the V-I behavior of SOFCs and SOFC stacks requires carrying out a suitable number of model runs for different pairs of the current density $i$ and the corresponding cell voltage $V_{cell}$ in order to cover the complete range of permissible operating points. This causes a great CPU time demand if all these simulations have to be performed using a 3D tool like the one presented in (1). The V-I curves are to be calculated dynamically as well so that the time needed to reach e.g. a new steady state after an instant load change...
can be obtained which increases the computational requirements even more. A previous study (2) using a 3D tool already dealt with an investigation similar to the one presented in this contribution but it was carried out for an electrolyte supported design that is obsolete now. In that previous study, an analytical relationship was developed based on dimensionless parameter groups for estimating the relaxation time (in (2) defined as the time the cell voltage needs to recover from an instant output current change to 90% of its new steady state value) for helping those engineers who do not have a sophisticated 3D code or simply do not benefit from it because it requires too much CPU time. This situation has now changed not only because cheap but powerful personal computers have become available today but also because of the recent development of a dynamic 1D code for co- and counterflow planar SOFCs. This code is simple and rapid enough to be attractive as a design tool for SOFC engineers if accuracy requirements are not too high.

MODEL ASSUMPTIONS AND INPUT VALUES

The 1D model's basic assumption is the possibility to focus on just one spatial coordinate, the x-direction parallel to the gas channels, because all the major concentration, temperature and current density changes occur in the direction of the gas flow. The width of the stack and its height can be neglected if an even flow distribution is ensured and the temperature deviations due to heat losses in the vicinity of the stack's outer walls can be neglected. An even flow distribution can be achieved e.g. by virtue of CFD-aided flow manifold design. The thermal deviations near the walls may be neglected if the stack is sufficiently large and the thermal insulation box the stack is placed in is of high quality.

The range of permissible operating points is given for the Jülich standard type stacks by the two constraints that the maximum temperature in the stack's solid body, mostly made up by the interconnector plates, or bipolar plates (BiP), \( T_{\text{bip}} \), must not exceed 800 °C and that \( V_{\text{cell}} \) must not drop below 0.6 V. Keeping \( T_{\text{bip}} \) below 800°C is deemed necessary to avoid accelerated stack degradation and limiting \( V_{\text{cell}} \) to 0.6 V is a precaution to prevent anode oxidation.

Simulations were performed for a Jülich standard type stack with anode substrate cells (20x20 cm²) with LSM cathodes (3). In all cases, the pressure is 0.1 MPa and the fuel is a gas mixture simulating pre-reformed natural gas with the following composition (in vol %):

| CH\(_4\) | H\(_2\)O | CO | CO\(_2\) | H\(_2\) | N\(_2\) |
|---|---|---|---|---|---|
| 16.6 | 39.7 | 1.0 | 8.2 | 33.3 | 1.2 |

The fuel enters at 500°C and the air in counterflow direction at 630°C in all cases. The absolute pressure is always 0.1 MPa. The electrochemical kinetics is approximated by a lump-sum internal resistance for cells with the current Jülich LSM cathodes with the following numerical values: \( \bar{R}_{\text{op}} = 0.29 \ \Omega \text{cm}^2 \) at \( T_{\text{op}} = 800 \ ^\circ\text{C} \) with \( E_A = 0.65 \ \text{eV} \). The corresponding equation is given below:
The internal resistance defined by the simplified approach according to equation [1] is very convenient because it can be easily obtained by evaluating measured V-I data from laboratory-type single cells at constant temperature and low fuel utilizations. The model used to describe the reforming kinetics had been presented in (4) already and had also been used in the previous study (2): A simple rate equation based on first order kinetics for the methane conversion followed by the shift equilibrium is therefore used in all the simulations presented in this contribution.

The chemical and electrochemical rate equations are needed as source terms in the conservation equations for mass- and heat flow required for describing the thermal behavior of the fuel stream, the air stream and the solid body. All this finally leads to a system of partial differential equations (PDEs) in the time \( t \) and one spatial coordinate \( x \) along the gas channels that must be solved numerically. In order to do that, boundary conditions are still needed. These are obtained by specifying zero heat flux in- and out of the stack’s solid body (adiabatic conditions) for all presented simulations, the inlet temperatures of the fuel and the air, the initial fuel composition and the current density \( i \). Hence the 1D tool’s job is to calculate reliable numerical approximations of the true solution of this system of PDEs for the specified boundary conditions.

As mentioned above, the model has adiabatic boundary conditions so heat transfer occurs only internally between the fuel and the BiP and the air and the BiP. The heat transfer is modeled by assuming a constant Nusselt number \( Nu = 3.66 \) because the flow is laminar in all cases. However the heat conductivities of the air and the fuel side must be known for calculating heat transfer coefficients. These heat conductivities are both composition and temperature dependent. The composition dependence on the air side is a minor effect in all the calculations discussed here because the air access number was quite high (5.25 times the stoichiometric minimum ratio needed to oxidize all the combustible fuel components). However, its knowledge is important on the fuel side because the fuel gas assumes various different compositions from an unreformed natural gas and steam mixture over a fresh reformate to an electrochemically utilized, depleted exhaust gas mixture. Fortunately, it can be shown that it is sufficient for the purpose of this work to use constant heat conductivities for the air and the fuel mixture determined by choosing representative values for a temperature range between 500°C and 800°C and fuel utilizations between 0% and 80%: On the air side, simply the value \( \lambda_{\text{air}} = 0.064 \text{ Wm}^{-1}\text{K}^{-1} \) for air at 650°C was chosen and on the fuel side, the value \( \lambda_{\text{fuel}} = 0.18 \text{ Wm}^{-1}\text{K}^{-1} \) for a fuel gas stemming from a CH\(_4\) and H\(_2\)O mixture with a H\(_2\O/\text{CH}_4\)-ratio (or Steam to Carbon ratio) of S/C = 2 at a fuel utilization of \( U_r = 40\% \) and 650°C. All these fluid heat conductivities were calculated using a previously in Julich developed computer code (5).

**SIMULATIONS**

As an initial step, a (stationary) base case was defined as a reference point for all the following variations: Figure 1 shows the temperature profiles of the bipolar plate and the gas streams (fuel and air in counterflow) as well as the corresponding mole fraction profiles of the fuel’s constituents. In this base case the stack is operated at an overall current den-
sity of \( i = 0.55 \text{ A/cm}^2 \). The fuel utilization is set at 80\% \( (U_F = 0.8) \). The total fuel flow rate is then calculated according to this current density and fuel utilization. The air flow (expressed in terms of \( \lambda \), which is the air excess number based on the total fresh fuel flow entering the SOFC), is used to limit the maximum temperature of the BiP to below 800 °C in order to avoid accelerated degradation. This results in a cell voltage (\( V_{\text{cell}} \)) of 0.7 V per cell on average for \( \lambda = 5.25 \).

The V-I curve simulations were divided in two cases: The first case refers to load changes introduced by altering the overall output current density \( i \) while the flow rates of fuel and air, respectively, are kept constant. Hence \( U_F \) will increase if \( i \) is increased and \( U_F \) will decrease if \( i \) is decreased. This way of load changing is often found in laboratory-type test operation of single cells and short stacks, for instance. The second case refers to load changes introduced by changing the overall output current density \( i \) while \( U_F \) is kept constant. This way of load changing is the preferred choice for operating technically and commercially interesting stacks and systems, for instance at part load, because there will be no unnecessary waste of fuel. \( U_F \) is defined by:

\[
U_F = \frac{i}{i_{\text{exhst}}},
\]

where \( i \) is the actual overall current density, for example in \text{A/cm}^2, and \( i_{\text{exhst}} \) is the exhaustion current density, also in \text{A/cm}^2, obtained after all combustible species in the fuel gas mixture will have been oxidized electrochemically so that the fuel is completely exhausted. It is therefore regarded as a more meaningful quantity for measuring the fuel flow per active stack area than the volumetric flow rate or the mole flow rate. Other quantities to be introduced here are the electrical power density \( P_{el} \) and the thermal efficiency \( \eta_{th} \):

\[
P_{el} = iV_{\text{cell}}
\]

\[
\eta_{th} = \frac{P_{el}}{(\dot{n}_F\Delta H)} = \frac{V_{\text{cell}}U_F}{V_{\Delta H}}
\]
In equation [4], \( \dot{n}_F' = i_{\text{ext}} / (zF) \) is the total mole flow rate per active cell area and stack plane of the combustible constituents of the fresh fuel entering the SOFC or the SOFC system (\( z \) is the number of charge transfers per elementary reaction, \( F \) is the Faraday constant). \( V_{\Delta H} = -\Delta H / (zF) \) is the voltage that would be measured if all the chemically stored energy, measurable as the enthalpy change \( \Delta H \) per mole of fuel at constant pressure and temperature, of the fuel could be exploited by the fuel cell process. It is assumed in all the here discussed simulations that the SOFC system runs on CH\(_4\) so the lower heating value at 25 °C is used yielding \( V_{\Delta H} = 1.02 \) V.

**Case 1: Constant Fuel and Air Flow, Variable Fuel Utilization**

Figure 2a) shows the temperature and concentration profiles for an idle case obtained from the base case by lowering the current output to almost zero (maintaining a small current was necessary for numerical reasons) at constant fuel and air flow so that \( U_F \) decreases to almost zero (1%). In addition, the fuel's mole fraction distribution is shown on the right in Figure 2b). Since there is no heat release due to the electrochemical oxidation, the temperature is dramatically lower as in the base case. The SOFC basically now acts as a reformer which temperature is mainly determined by the heat supplied to the reforming process from the air entering on the right which is then cooled down by the reforming process. The mole fraction profile shows that the reforming process approaches its chemical equilibrium at about 625°C but won't be able to achieve it because the gas channel is too short for the low reforming rate at this low temperature.

Figure 2. Temperature (a) and concentration (b) profiles for the idle operation (\( i \approx 0 \)) (case 1: fuel and air flow rates constant).

Figure 3a) shows the cell voltage as well as \( U_F, P_{\text{el}} \) and \( \eta_{\text{th}} \) as a function of \( i \). These curves were obtained by calculating several stationary operating points at different current densities \( i \) while the fuel and the air flow were left unchanged. It is interesting to note that the non-linear shape of the V-I curve is mainly caused by the temperature increase because a current- and partial pressure independent internal resistance was used. In Figure 3b) on the right, there is the corresponding maximum temperature in the bipolar plate. At \( i < 0.2 \) A/cm\(^2\), the reforming reaction consumes only slightly less heat than the electro-
chemical oxidation releases. So the stack’s temperature response to a current increase is somewhat sluggish at low current densities.

Figure 3. V-I Curve for case 1: (a) $V_{cell}$, $U_F$, $\eta_{th}$, $P_{el}$ and (b) $T^{\text{max}}$ as $f(i)$.

Figure 4 shows the transient development of the BiP’s temperature profile from a steady state at 0.15 A/cm² after an instant load change (current increase) to a new steady state at 0.35 A/cm² and from there on back to the base case at 0.55 A/cm². As mentioned above, the fuel and the air flow were constant. After the first load change, it takes about 45 min to reach the new steady state at 0.35 A/cm² and subsequently about 35 min to re-establish the base case. If the current draw is instantly increased by 0.4 A/cm² all at once (not shown in Figure 4) from 0.15 A/cm² to the base case skipping the intermediate operating point, $V_{cell}$ drops below 0.3V.

Figure 4. Temperature profiles after transient load changes at constant flow rates.
Although simulations show that $V_{\text{cell}}$ then gradually recovers to the base case value at 0.70 V over time, this is not tolerable since $V_{\text{cell}} < 0.6$ V may damage the anode due to Ni oxidation. Hence smaller steps must be chosen. In this case, steps of 0.2 A/cm$^2$ at a time kept the initial $V_{\text{cell}}$ values in the range of 0.55 to 0.6 V, so this seems the maximum tolerable ramp-up step size for the current output here.

It is also important to note in Figure 4 that the position of the temperature maximum shifts from the air inlet toward the fuel inlet side with increasing $i$. This may constitute a problem in monitoring the maximum temperature during real SOFC operation since this parameter is, amongst others, used to control the operation of the system.

Figure 5a) shows the evolution of the temperature profile over time for the second load change from 0.35 A/cm$^2$ back to the base case. The response of $V_{\text{cell}}$ is shown on the right in Figure 5b). The cell voltage plunges instantly from 0.73 V down to 0.57 V at $t = 0$ before it recovers to a new steady state, the base case, and eventually stabilizes at 0.70 V. The initial plunge to below the steady state value of 0.70 V at 0.55 A/cm$^2$ is due to the lower temperature level at 0.35 A/cm$^2$. After the current increase, more heat is released and so the temperature profile starts rising. As a result, the internal resistance decreases causing $V_{\text{cell}}$ to increase again until a new steady state heat balance is established and the temperatures do not change any more.

Figure 5. Evolution of (a) $T_{\text{HP}(x,t)}$ and (b) $V_{\text{cell}}(t)$ after a current increase (case 1).

Figure 6a) and b) are similar figures showing the transition from the base case to the idle point. After a current shut-off at constant gas flow rate, the cell voltage jumps up instantly from 0.70 V to approx. 1 V at $t = 0$ and then it starts drifting up further to a steady state at about 1.02 V. The cell voltage is governed by the Nernst potential at idle operation (OCV) which increases with decreasing temperatures. After the power shut down, the stack needs about half an hour to cool down which is due to the still continuing internal reforming process.
Figure 6. Cool-down to the idle point after a power shut-down (case 1): a) $T_{\text{BIP}}(x,t)$ and b) $V_{\text{cell}}(t)$.

**Case 2: Constant Fuel Utilization, Variable Fuel and Air Flow**

Figure 7a) shows the idle point at constant fuel utilization. $U_F = 0.80$ was maintained in this and the other calculations for case 2. Also, $\lambda$ was kept constant and so the fuel flow rate was always changed together with the air flow rate. For the idle case, a small current density of 0.05 A/cm$^2$ had to be taken for numerical reasons. Maintaining a constant $U_F$ requires reducing the fuel flow to achieve part load operation at lower current densities. The reduced velocities increase the contact time with the anode catalyst and so both the reforming process and the electrochemical oxidation occur mostly within the first 20% of the gas channel close to the fuel inlet as shown in Figure 7b).

![Temperature profile](image1)

**Figure 7. Idle point for case 2: a) T-profiles and b) mole fraction profiles.**

The temperature profile in Figure 7a) still shows a flat maximum and it is higher than in case 1. The flat temperature profile is due to a higher ratio of heat conducted in the solid body to the net heat release given by the difference between heat released through the electrochemical conversion and the heat consumed through the reforming which results in low temperature gradients in the solid body. The much lower level of the temperature profile as a whole compared with the base case is a result of the low current density that
diminishes the heat release rate even though $U_F$ is still 80%. $U_F = 0.8$ also explains why the temperature profile settles at a higher level in case 2 than in case 1, see Figure 2a) as well: Although the current density is very low, still 80% of the fuel are oxidized because the gas flow is very low either which causes a temperature increase not seen in Figure 2a). Since there is almost no fuel utilization, the idle point of case one is completely governed by the reforming process.

Figure 8a) then shows the V-I curve for case 2 on the left, along with $U_F$, $\eta_{th}$, and $P_{el}$. The shape of the V-I curve is different compared with case 1 as seen in Figure 3a). The reason is seen on the right where the corresponding maximum of $T_{Bip}$ is displayed: This function has a negative curvature (2nd derivative) while Figure 3b) shows a positive one. That different behavior is again explained by the heat balance between the heat release rate and the heat transport in the bipolar plate: The reforming reaction cannot be as dominant as in case 1 because there is always $U_F = 80\%$ so there is always some heat production as long as there is a finite current draw even though the gas flow is very low to accommodate the high fuel utilization.

Figure 9 shows three exemplary temperature profiles (compare with Figure 4 for case 1 as well) which maxima correspond with Figure 8. The temperature profiles for case 1 and these for case 2 look similar but there are differences: The most important difference is that the temperature maxima of the profiles for the 3 different current densities stay much closer to each other as in case 1: They are all located between 30% and 40% of the gas channel length. This fact makes it lot easier to prepare the stack for monitoring the temperature maximum at $U_F$ = const. than in case of constant gas flow as in case 1 because thermocouples are needed only between 30% and 40% of the gas channel length and not across the entire bipolar plate.

Figure 10 shows the transient load change after an instant current increase covering the full range from the idle point all the way back to the base case from 0.05 A/cm² to 0.55 A/cm² skipping the immediate operating points at 0.15 A/cm² and 0.35 A/cm² shown in Figure 9. Just like in the figures discussed before, an immediate cell voltage drop at $t = 0$ from 0.92 V down to 0.55 V is initiated by the instant current jump. Although $V_{cell}$ falls to 0.55 V, which is below the threshold at 0.6 V, it then recovers fairly quickly and the new steady state at 0.70 V is reached within one hour. The reason for the initial voltage drop

**Figure 8. V-I Curve for case 2: a) $V_{cell}$, $U_F$, $\eta_{th}$, and $P_{el}$ and b) $T_{max}$ as f($i$).**

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Figure 9. Temperature profiles after transient load changes at constant $U_p$.

is the same as in case 1 (see also Figure 5): The increased current density causes a voltage plunge below the new steady state because of the higher resistance at the lower temperatures. On the other hand, the larger current draw also causes an increased resistive heating so the temperatures rise again, the internal resistance decreases as a consequence so $V_{cell}$ starts moving up again to its new steady state at 0.70 V. It is interesting that contrary to case one, see Figure 4 and its discussion, it may be possible to ramp up the current in a single step from the idle point to the base case when operating the SOFC at constant fuel utilization and not at constant fuel and air flowrate. However, as a precaution, two current steps could be taken instead of only one. Simulations (not displayed in the figures) show that a current increase from 0.05 A/cm$^2$ to 0.15 A/cm$^2$ and then to the base case at 0.55 A/cm$^2$ keeps $V_{cell}$ above the threshold at 0.6 V.

It may also be interesting to note that there is an initial temperature drop at $t = 0$ and $x = 0$ (fuel inlet) and $x = 1$ (air inlet) while the rest of the profile is moving up to higher temperatures from the start already. This normalizes after approx. 5 min when the entire temperature profile starts rising at all locations.

Figure 10. Evolution of $T_{BIP}$ and $V_{cell}$ after a current increase (Case 2):
a) $T_{BIP}(x,t)$ and b) $V_{cell}(t)$. 
That initial temperature drop is explained by the increase in fuel and air flow causing an additional cooling effect by the reforming process at the fuel inlet and by the increased heat transfer to the air at the air inlet. At the same time, the heat release rate is greater due to the increased current density so the temperature starts rising in the middle of the BiP while it drops at the endpoints. This goes on for about five minutes until the temperature gradients are steep enough to transport sufficient heat to both gas inlets via solid body conduction to compensate that initial drop.

Figure 11 shows the reverse process to the one shown in Figure 10 which is the transient cool down process from the base case to the idle point after an instant current decrease from 0.55 A/cm² down to 0.05 A/cm². At both ends of the BiP, there is an initial temperature increase instead of a drop. That is caused by a local heat load reduction due to less cooling by the reforming and the air as a result of the gas flow reduction that had to be done simultaneous to the current reduction. It is remarkable that the entire cool down process stretches out over more than ten hours before the cell voltage is finally stable again and the idle point is reached. As in case 1 (see Figure 6), \( V_{\text{cell}} \) instantly leaps up to just above 0.9 V. Contrary to the situation shown in Figure 6 the voltage decreases a little bit before it starts increasing again to a new steady state at about 0.92 V. The first decrease is caused by the rapid decline of the temperature maximum due to less resistive heating in which vicinity most of the current is generated. The simultaneous temperature increase at the fuel and the air inlet is less important for the current production at this stage since it is still too cold there. After about 30 min, the entire temperature profile starts moving down but now both ends of the BiP are much hotter so the impact of the maximum on the resistance becomes less important and the current production is more evenly distributed over the entire active area. That causes the overall (average) internal resistance to decrease again so the voltage drop due to the current flow is just about low enough to allow the Nernst potential to control \( V_{\text{cell}} \) which starts increasing again from now on. The long time span necessary to reach the new steady state can now be explained by only a very slight dominance of the Nernst potential over the resistive voltage drop during the entire duration of the transition.
CONCLUSIONS

There is a strong influence of the temperature on the cell voltage at given current density as can be expected. The maximum current density that can be drawn from the stack must be counterbalanced by an appropriate air flow to cool the stack so that a certain maximum temperature of the stack of 800 °C is not exceeded to avoid accelerated degradation. However, the internal resistance increases with decreasing temperature so care must be taken that the cell voltage does not drop below the potential for Ni oxidation at about 0.6 V at high current densities.

All transient load changes show an instant voltage jump as a result of an instant current change. Since the thermal balance is changed as well, the voltage may undershoot or overshoot at first before it then starts following the temperature profile drift toward a new steady state. Stepping up the current at constant fuel utilization shows a more stable dynamic SOFC behavior than at constant gas flow rates (fuel and air). It is possible to turn up the current e.g. from 0.05 A/cm² to 0.55 mA/cm² without causing an initial voltage drop much below 0.6 V at constant fuel utilization. If the current is increased in two steps from 0.05 A/cm² to 0.15 A/cm² and then to the base case at 0.55 A/cm², $V_{\text{cell}}$ can be kept above the threshold at 0.6 V while the current steps must be limited to max. 0.2 A/cm² at constant gas flow rates. On the other hand, an instant power shut down seems possible in both cases because the cell voltage rises immediately following the power shut-down instead of falling. Transient current load changes at constant fuel and air flow tend to reach a new steady state more rapidly than corresponding ones at constant fuel utilization. Especially an instant power shut-down at constant $U_F$ takes hours before a stable idle point is reached. Stepping up the current requires between half an hour to 45 min at constant gas flow rate and up to about 1.3 hours at constant fuel utilization to reach a new steady state for the standard Jülich design.

The occurrence of a temperature maximum in the solid body of a planar SOFC operating in counterflow mode for the fuel and the air and utilizing hydrocarbon fuel with internal reforming is typical. It is important to monitor the temperature maximum of the BiP because that is essential to avoid local overheating in case of internal reforming. Since this maximum stays more or less in one place during technically interesting load changes at constant fuel utilization, it may be possible to measure the maximum temperature with a few thermocouples in the BiP. However, if the stack was to be operated at constant gas flow rates, the maximum temperature would sweep from the air inlet across the bipolar plate toward the fuel inlet which would make it necessary to cover the entire active area with enough thermocouples to pick up the maximum temperature reliably.

Future work in the area of dynamic SOFC modeling should include the development of a suitable SOFC system control strategy for handling these (sometimes quite long) load change durations.

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