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Spatial Distribution of Electrochemical Performance in a Segmented SOFC: A Combined Modeling and Experimental Study

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Spatial Distribution of Electrochemical Performance in a Segmented SOFC: A Combined Modeling and Experimental Study

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Abstract
Spatially inhomogeneous distribution of current density and temperature in solid oxide fuel cells (SOFC) contributes to accelerated electrode degradation, thermomechanical stresses, and reduced efficiency. This paper presents a combined experimental and modeling study of the distributed electrochemical performance of a planar SOFC. Experimental data were obtained using a segmented cell setup that allows the measurement of local current-voltage characteristics, gas composition and temperature in 4×4 segments. Simulations were performed using a two-dimensional elementary kinetic model that represents the experimental setup in a detailed way. Excellent agreement between model and experiment was obtained for both global and local performance over all investigated operating conditions under varying H₂/H₂O/N₂ compositions at the anode, O₂/N₂ compositions at the cathode, temperature, and fuel utilization. A strong variation of the electrochemical performance along the flow path was observed when the cell was operated at high fuel utilization. The simulations predict a considerable gradient of gas-phase concentrations along the fuel channel and through the thickness of the porous anode, while the gradients are lower at the cathode side. The anode dominates polarization losses. The cell may operate locally in critical operating conditions (low H₂/H₂O ratios, low local segment voltage) without notably affecting globally observed electrochemical behavior.

Keywords: Solid oxide fuel cell (SOFC), Segmented cell, Experiment, Modeling, Simulation

1 Introduction
High electrical efficiency and long-term durability are key requirements for a cost-competitive introduction of fuel cell technology. In order to achieve high efficiency, a high fuel utilization is required, that is, the complete fuel should be converted electrochemically in the fuel cell. In solid oxide fuel cells (SOFC), high fuel utilization results in strong concentration gradients at the anode, where the fuel (H₂, CO, CH₄) is successively diluted by reaction products (H₂O, CO₂). This, in turn, leads to an inhomogeneous distribution of electrochemical performance (local power density) and temperature. These inhomogeneities detrimentally affect both efficiency (through a large concentration polarization) and long-term stability (through thermomechanical stresses and local operating conditions that may induce degradation). In order to understand these effects, it is necessary to obtain detailed information about the spatial distribution of the electrical, chemical and thermal cell properties.
For polymer electrolyte fuel cells (PEFCs), segmented cell setups for the local measurement of current density have been developed and are widely used.

In contrast to this, only few experiments for a spatially resolved characterization of SOFCs have been conducted so far, mainly because the high operating temperature (700-900 °C) represents a considerable challenge to the experimental setup. One of the first setups for the local characterization of segmented planar SOFC cells has been developed by Metzger et al.

Segmented-cell setups are typical model experiments in the sense that they allow a largely extended insight at the cost of modified geometries and/or materials in comparison to standard fuel cells. Results from segmented cells are therefore not expected to be quantitatively equivalent to those from standard stacks in terms of performance or temperature distribution. Having this difference in mind, the accessible information is highly valuable both for increasing the fundamental understanding and for optimizing cell and flow field design.

The use of mathematical modeling in combination with experimental investigations can significantly enhance the understanding of the spatially resolved electrochemical behavior of the cell. For example, today’s experimental techniques do not allow the measurement of concentration and current distributions inside the porous electrodes. However, it is the electrochemically active zone inside the electrodes that governs electrochemical performance as well as degradation behavior. For this reason, modeling studies are increasingly used to aid understanding of the fuel cell processes and to optimize electrode and cell design.

The combination of simulations with segmented-cell experiments is particularly useful because it allows reliable model validation including local behavior. Furthermore, the segmentation can easily be omitted and geometrical parameters be modified in the model, thus allowing to bridge the difference between the segmented cells and standard cells.

In this paper we present a combined modeling and experimental study of the spatial distribution of electrochemical performance in a planar SOFC. The experiments are based on the setup by Metzger et al. The simulations are conducted within a physically-based modeling framework developed by Bessler et al., which is extended in order to reflect the segmentation of the experimental setup. The following approach is used: (i) The model is validated using experimental polarization data under specifically designed calibration conditions; (ii) model predictions are compared with experimental data at realistic operating conditions; and (iii) the validated model is used for quantitative interpretation and prediction of the physical, electrochemical and fluid mechanical fuel cell properties.

2 Experimental Setup

The experimental setup for segmented cells has been described previously.

It is shown schematically in Fig. 1a. Square-shaped anode-supported cells (100 cm² area, 540 µm Ni/YSZ cermet anode with a thin anode functional layer, 5 µm 8YSZ electrolyte, 3 µm YDC interlayer, 45 µm LSCF cathode divided into 16 segments with an active area of 4.84 cm² of each segment) were integrated in a metallic cell housing and sealed with glass seal (Fraunhofer Institute of Ceramic Technologies and Sintered Materials (IKTS), Dresden, Germany). The metallic housing is subdivided into 16 galvanically isolated segments with a total active area of 73.96 cm². The flow field is identical for fuel and air side; it consists of square-shaped channels (2 x 2 mm, 5 channels per segment row) that are supplied via inlet and outlet manifolds that are symmetrical to each other (ca. 4 cm² per manifold).

In order to determine the temperature at each segment, thermocouples are introduced in the metallic segments. Additionally, capillary tubes that correspond to the cathodic segments are integrated at the anode side at 16 measuring points to take samples of the anode gas to be analyzed by gas chromatography. The setup allows for the integral and spatially resolved measurement of current density and voltage, the local and integral determination of impedance data, the local measurement of temperature and the spatially resolved analysis of the fuel gas concentrations along the flow path.

Experiments were conducted under a wide range of operating conditions as summarized in Table 1. Conditions for model calibration were defined that show a high sensitivity of simulated polarization curves on unknown model parameters. The calibration measurements were carried out under high gas flow rates (low fuel utilization) in order to avoid strong fuel depletion. Even for the highest fuel dilution (Table 1, condition 6), fuel utilization was < 26 %. Additional measurements were performed under realistic gas compositions at high fuel utilization (Table 1, condition 10).

3 Modeling and Simulation

3.1 Planar SOFC Modeling Framework

The framework for physically-based SOFC modeling and simulation has been presented previously. The model layout is shown schematically in Fig. 2, and all model parameters are summarized in Table 2. A detailed 1D+1D elementary kinetic electrochemical model was applied that represents one single channel of the experimental setup (Fig. 2b). One-dimensional channel flow (x dimension) is described using the Navier-Stokes conservation equations (continuity, species, momentum), corresponding essentially to a plug-flow model with axial diffusion. One-dimensional mass transport through the membrane-electrode assembly (MEA) (y dimension) is described by coupled Fickian/Knudsen diffusion and Darcy flow. Charge transport in the solid...
electrolyte and the electrolyte phase of the composite electrodes is described in two dimensions using Ohm’s law.

The results shown in this paper are calculated under the assumption of isothermal conditions. This assumption is justified because measured temperature variations along the flow path are relatively low (26 K at a current density of 1.4 A/cm²) due to rapid heat losses via the metallic flow field and housing.

Anode electrochemistry is described using elementary kinetics for surface reactions on Ni and YSZ, and for charge transfer (hydrogen spillover) (Fig. 2c), as given in Table 3, and as validated by previous cermet anode and patterned anode studies. The electrochemical double layer is described as simple capacitor. Cathode electrochemistry is described using a modified Butler-Volmer equation,

\[ i = i^0_p \exp(-E_{act}/RT) \cdot [ \exp(0.5F\eta/RT) – \exp(-0.5F\eta/RT) ] . \] (1)

A definition of all symbols and their units is given in 13. Current-voltage relationships are modeled by directly solving for the electric-potential distribution in the electrodes and electrolytes without using the Nernst equation. All models are implemented in the in-house numerical simulation software DENIS. For numerical simulation, the MEA was spatially discretized into 16 x 80 control volumes (x x y dimension). For identifying unknown model parameters, a Levenberg-Marquardt nonlinear least-squares fitting algorithm was used 17.

3.2 Segmentation

For the present study, the model described in Ref. 13 was extended in order to include the cell segmentation of the experimental setup. The segmentation is represented by an electrical circuit model as shown in Fig. 2a. An arbitrary number and size of segments can be used at both anode and cathode. The segments are individually connected via switches S_i to outer power rails at anode and cathode side. These rails are assumed to be at constant potential \( \phi \) given by the outer load. The connection wires are assumed to be resistors R, leading to a potential drop modeled via Ohm’s law,

\[ \Delta \phi = R_i \cdot \Delta \overline{\phi} \] (2)

where \( R \) is the cell-area-specific wire resistance and the potential drop \( \Delta \phi \) depends on the segment current density \( \Delta \overline{\phi} \). Along the length of each individual segment the potential and therefore the local segment voltage \( U_{local \_i} \) are assumed constant. The simulation input is the voltage \( U \) of the external load. The model yields the total current \( I \), the individual segment currents \( I_i \), as well as local segment voltages \( U_{local \_i} \). The latter are determined experimentally via individual voltage probes (four-point measurements at each segment). The model can also be used to calculate global and local impedance \( Z \) and \( Z_{local} \), respectively.

For the present simulations, four segments are used at both anode and cathode, and all switches \( S_i \) are closed, representing the experimental setup. The wire resistance \( R \) is determined via fitting to the experiments (cf. Table 2).

4 Results and Discussion

4.1 Calibration Measurements and Model Validation

There are a number of model parameters associated with electrochemistry and transport in the MEA that are specific for the investigated cells and cannot be obtained from literature. Some parameters can be reliably estimated (e.g., porosity from SEM micrographs). Other parameters, however, can only be obtained by fitting to experimental data. To this goal, the model was validated under specifically designed operating conditions (Table 1, conditions 1-9). Experimental polarization behavior was measured under low fuel utilization (< 15 %) by using fast gas inflow velocities in order to circumvent spatial gradients along the channel. Gas-phase anode and cathode inlet concentrations as well as temperature were varied over a range of conditions; these conditions were chosen in order to achieve sensitivity for the unknown parameters. Parameters were fitted using a one-dimensional model of the MEA only. Fitting results are included in Table 2 and Table 3.

In the present study, the following model parameters were fitted: (1) The preexponential factor of the elementary kinetic hydrogen spillover reaction at the anode (all other elementary kinetic parameters were used without modification from previous cermet anode studies); (2) the exchange current density of the global kinetic charge-transfer reaction at the cathode; (3) the \( p_{o2} \) exponent for cathode; (4) the tortuosity of the anode; (5) the ohmic resistance of the current collector wires. Given the high level of physicochemical detail of the present model, the number of free fit parameter has to be considered very low. It should be noted that the preexponential factor of the anodic charge-transfer reaction had to be modified by only 30 % in comparison to previous cermet anode studies; this small modification can be interpreted as variations in material and/or microstructure and demonstrates how elementary kinetic approaches can reduce empiricism in fuel cell modeling.
Simulated and experimental polarization curves for the validation conditions are shown in Fig. 3. There is good quantitative agreement between model and experiment over the complete range of operating conditions (anode H₂/H₂O ratio, anode gas dilution with N₂, cathode O₂/N₂ ratio, and temperature). Note that the physical model uses only one single set of parameters for all conditions. A systematic deviation between model and experiment occurs during temperature variation. We believe that this is due to an onset of cell degradation, lowering the experimentally observed performance relative to the model predictions. Degradation effects are not included in the model.

4.2 Measurement and Simulation Under High Fuel Utilization

The segmented cell was operated under a condition with high fuel utilization (Table 1, condition 10). The anode was fed with 33% H₂, 1% H₂O and 66% N₂. This condition was chosen in order to simulate nitrogen-rich reformate gas. The cathode was fed with air in counter-flow operation.

The measured two-dimensional distribution of power density in the 16 segments is shown in Fig. 4 for an average power density of 460 mW/cm². The fuel utilization at this condition is 80%. The cell performance is strongly inhomogeneous, with the power density systematically decreasing from fuel inlet (left side of Fig. 4) to fuel outlet (right side of Fig. 4). It will be shown below that this decrease is due to fuel depletion along the flow path. Moreover, there is a notable difference for the four segment rows (upper row to lower row in Fig. 4). This difference may be due to sealing issues or inhomogeneous gas supply to the gas channels. Further segment-to-row variations are likely due to a variation in contact resistance, which may also lead to systematic row-to-row variations when the contact pressure is inhomogeneous. For comparison with the model (which represents the behavior across an entire channel), the row with segments 9-12 was chosen.

Experimental and simulated global and local current-voltage characteristics for segments 9-12 are shown in Fig. 5. Simulations were performed using the 2D model (i.e., along one representative channel and through the thickness of the MEA, cf. Fig. 2). The global IV-curve (Fig. 5a) shows a typical shape with a parabolic behavior at low currents, linear behavior at intermediate currents, and a limiting current density of ~0.8 A/cm² at high currents. The maximum power density (P_{max} = 470 mW/cm²) is observed at a cell voltage of 0.70 V. Local IV-curves for segments 9-12 are represented by plotting local segment voltage versus local segment current (Fig. 5b). The local behavior shows a considerable variation of current density for different cell segments. At high polarization, segments 11 and 12 that are located close to the fuel outlet show a particularly interesting behavior: the current decreases while at the same time the segment voltage strongly decreases. This "inverse" behavior is due to strong fuel depletion (see next section). At the same time, the current density of segment 9 that is located at the fuel inlet continues to increase.

There is excellent quantitative agreement between model and experiment for both the local and the global behavior. Note that no additional parameter adjustment was performed after model calibration (Section 4.1). This finding also underlines the validity of the choice of calibration conditions.

4.3 Distribution of Gas-Phase Concentrations

Fig. 6 shows the simulated spatially resolved distribution of gas-phase concentrations in the channels as well as in the porous electrodes and current collector meshes for the condition of maximum power density (P_{max} in Fig. 5a). There are considerable concentration gradients throughout the complete system. In particular, the relatively thick porous anode in this anode-supported cell is responsible for strong H₂ depletion through the anode thickness. This is the case, in particular, for segments 11 and 12. Here, H₂ concentration tends towards zero at the porous electrode/solid electrolyte interface. This is the reason for the strong decrease in segment voltage observed in the local IV-curves (Fig. 5b).

The upper panel of Fig. 6 also shows the local current density along the flow path. The step-like behavior of the four segments is due to the contribution of the resistance of current collector wires (cf. Fig. 2a): The potential drop over the wire for the individual segments decreases as the overall segment current decreases along the fuel flow path (left to right in Fig. 6). This leads to an increase in integral segment current, causing a step from the end of one segment to the beginning of the next segment. Note that the segment voltage is measured and simulated directly at the segment (U_{local} in Fig. 2a), therefore including the wire resistance. The current density distribution along the length of each segment is inhomogeneous. The integral current density over one segment is shown in Fig. 5b.

The condition of strong fuel depletion observed inside the porous anode may be highly critical concerning cell degradation. In particular, low H₂/H₂O ratios may lead to nickel oxidation at the anode, and low local segment voltages may lead to cathode degradation at segment 12 is not obvious from the global IV-behavior. Thus, a cell may be locally in a critical operating condition while global characteristics are well within apparent stability limits (e.g., sufficiently high global cell voltage). Post-test analysis and interpretation with a focus on degradation is subject of ongoing studies.

4.4 Distribution of Polarization Losses
The knowledge of the physical origin of polarization losses is a requirement for an optimization of cell design and operating conditions. The validated model offers the possibility to quantify the spatially varying polarization losses. Results for segments 9 and 12 are shown in Fig. 7 for the same operating conditions as shown in Fig. 5. The total polarization losses are separated into contributions from Ohmic resistance of the electrolyte \( \eta_{\text{ohm}} \), concentration polarization \( \eta_{\text{conc}} \) at both anode and cathode, and activation polarization \( \eta_{\text{act}} \) at both anode and cathode.

For both segments, concentration polarization at the anode is the dominant loss process. This finding is consistent with the observation of strong H\(_2\) depletion along the channel and through the anode thickness. The second largest contribution is anode activation. Polarization due to the electrolyte and cathode activation and concentration losses are low and do not significantly affect the cell performance. With increasing current density, all losses increase up to the point of maximum power density. Beyond that point, Ohmic and activation losses decrease in segment 12 due to the decreasing local current density in these segments (cf. Fig. 5b).

The analysis shows that the parabolic behavior at low current density that is observed in the global IV-curve (Fig. 5a) is due to anode concentration polarization and not, as commonly assumed, due to activation polarization. This has been pointed out before \(^{14}\). In this respect, SOFCs behave differently from PEFCs where low-current parabolic behavior is indeed due to activation losses.

5 Conclusions
A combined experimental and modeling study of the spatial distribution of the electrochemical performance in a planar SOFC was performed. Experimental data were obtained using a specifically designed segmented cell setup that allows for the measurement of local current-voltage characteristics, gas composition and temperature. Simulations were performed using a two-dimensional elementary kinetic model that represents the experimental setup in a highly detailed way.

Model parameters were identified by comparing simulations with validation experiments under low fuel utilization. Experiments and simulations under high fuel utilization showed strong gradients of gas concentrations and current density along the flow path and throughout the thickness of the membrane-electrode assembly. Over the complete range of operating conditions, excellent quantitative agreement between model and experiment for both global and local behavior was observed. The relatively thick porous anode and high fuel utilization causes particularly strong concentration variations at the anode, while the gradients are lower at the cathode side. The combined modeling and experimental study allows for the reliable quantification of the contribution of various polarization losses. It was shown that concentration polarization at the anode is the dominant loss process, followed by anode activation polarization. The anode was therefore identified as the critical component in this cell type.

An important conclusion is that the cell may operate locally in critical operating conditions (low H\(_2\)/H\(_2\)O ratios, low local segment voltage) without notably affecting global electrochemical behavior. These conditions are likely to detrimentally affect cell durability. Based on a combination of experiment and modeling, the present study allows for identifying such conditions and for assessing optimized cell design and operation strategies.

Acknowledgements
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References


### Figure Captions

Fig. 1: Setup of measurement system for the characterization of planar segmented SOFCs with cathode segmentation for anode-supported cells.

Fig. 2: Model of the segmented SOFC. (a) Representation of segmentation by an electrical circuit. (b) Two-dimensional representation of one single gas channel including MEA and interconnect. (c) Surface electrochemistry of H$_2$ oxidation at the anode three-phase boundary.

Fig. 3: Comparison of experimental data under validation conditions (low fuel utilization) and 1D model calculations. (a) Variation of anode gas composition without N$_2$ at 800 °C with cathode at 100 % O$_2$; (b) Variation of anode gas dilution with 50 % H$_2$ and 50 % H$_2$O at 800 °C and cathode at 100 % O$_2$; (c) Variation of cathode gas composition at 800 °C with anode at 50 % H$_2$ and 50 % H$_2$O; (d) Variation of temperature with anode at 50 % H$_2$ and 50 % H$_2$O and cathode air.

Fig. 4: Measured two-dimensional distribution of power density over the 16 segments under operating conditions with high fuel utilization (counter-flow operation; anode: 33 % H$_2$, 1 % H$_2$O, 66 % N$_2$ in H$_2$, 1.1 m/s inflow velocity; cathode: air, 5.2 m/s inflow velocity; $T = 800$ °C) at a cell voltage of 0.59 V. The fuel inlet is at the left side, the air inlet at the right side.

Fig. 5: Experimental and simulated polarization behavior for the segmented cell using the 2D model under operating conditions with high fuel utilization (counter-flow operation; anode: 33 % H$_2$, 1 % H$_2$O, 66 % N$_2$ in H$_2$, 1.1 m/s inflow velocity; cathode: air, 5.2 m/s inflow velocity; $T = 800$ °C). (a) Global current-voltage curves, (b) Local segment voltage versus local segment current. The numbers indicate the segments, where segment 9 is the first in flow direction of the fuel gas.

Fig. 6: Distribution of local current density and gas-phase concentrations within channel and porous electrodes at anode and cathode side for the point of maximum power density $P_{\text{max}}$ (cf. Fig. 5). The numbers in the upper panel indicate the segments.

Fig. 7: Calculated contributions of polarization losses in segments 9 and 12 in dependence on cell voltage for the same operating conditions as shown in Fig. 5. The broken lines represent the condition of maximum power density (Fig. 6).
Table 1: Experimental measurement conditions. Conditions 1-9 represent calibration conditions (low fuel utilization and specific N<sub>2</sub> dilution), condition 10 a realistic operating condition (high fuel utilization).

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Table 2: Model parameters used for all calculations.

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<td>5.15·10&lt;sup&gt;7&lt;/sup&gt; SK/m, 84 kJ/mol</td>
<td>Micro-structural model&lt;sup&gt;15&lt;/sup&gt;</td>
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<td>Resistance of wires</td>
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<td>Exp.</td>
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<td><strong>Conditions</strong></td>
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<td>Temperature</td>
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<td>Pressure</td>
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<td>Exp.</td>
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</table>

<sup>a</sup> Estimated.
<sup>b</sup> Fitted to the present experiments.
Table 3: Elementary kinetic reaction mechanism for the anode and global kinetic reaction for the cathode. A single hydrogen spillover between Ni and YSZ surfaces is assumed as active charge-transfer reaction with a symmetry factor $\alpha = 0.5$. The surface site densities of Ni and YSZ are $6.1 \times 10^{-9}$ and $1.3 \times 10^{-9}$ mol/cm$^2$, respectively.
Fig. 1: Setup of measurement system for the characterization of planar segmented SOFCs with cathode segmentation for anode-supported cells.

150x210mm (600 x 600 DPI)
Fig. 2: Model of the segmented SOFC. (a) Representation of segmentation by an electrical circuit. (b) Two-dimensional representation of one single gas channel including MEA and interconnect. (c) Surface electrochemistry of H2 oxidation at the anode three-phase boundary.

209x297mm (600 x 600 DPI)
Fig. 3: Comparison of experimental data under validation conditions (low fuel utilization) and 1D model calculations. (a) Variation of anode gas composition without N2 at 800 °C with cathode at 100 % O2; (b) Variation of anode gas dilution with 50 % H2 and 50 % H2O at 800 °C and cathode at 100 % O2; (c) Variation of cathode gas composition at 800 °C with anode at 50 % H2 and 50 % H2O; (d) Variation of temperature with anode at 50 % H2 and 50 % H2O and cathode air.
Fig. 4: Measured two-dimensional distribution of power density over the 16 segments under operating conditions with high fuel utilization (counter-flow operation; anode: 33 % H₂, 1 % H₂O, 66 % N₂ in H₂, 1.1 m/s inflow velocity; cathode: air, 5.2 m/s inflow velocity; T = 800 °C) at a cell voltage of 0.59 V. The fuel inlet is on the left side, the air inlet at the right side.
Fig. 5: Experimental and simulated polarization behavior for the segmented cell using the full 2D model under operating conditions with high fuel utilization (counter-flow operation; anode: 33 % \( \text{H}_2 \), 1 % \( \text{H}_2\text{O} \), 66 % \( \text{N}_2 \) in \( \text{H}_2 \), 1.1 m/s inflow velocity; cathode: air, 5.2 m/s inflow velocity; \( T = 800 \, ^\circ\text{C} \)). (a) Global current-voltage curves. (b) Local segment voltage versus local segment current. The numbers indicate the segments, where segment 9 is the first in flow direction of the fuel gas.
Fig. 6: Distribution of local current density and gas-phase concentrations within channel and porous electrodes on anode and cathode side for the point of maximum power density $P_{\text{max}}$ (cf. Fig. 5). The numbers in the upper panel indicate the segments.

169x206mm (600 x 600 DPI)
Fig. 7: Calculated contributions of polarization losses in segments 9 and 12 in dependence on cell voltage for the same operating conditions as shown in Fig. 5. The broken lines represent the condition of maximum power density (Fig. 6).
Spatial Distribution of Electrochemical Performance in a Segmented SOFC: A Combined Modeling and Experimental Study

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Abstract
Spatially inhomogeneous distribution of current density and temperature in solid oxide fuel cells (SOFC) contributes to accelerated electrode degradation, thermomechanical stresses, and reduced efficiency. This paper presents a combined experimental and modeling study of the distributed electrochemical performance of a planar SOFC. Experimental data were obtained using a segmented cell setup that allows the measurement of local current-voltage characteristics, gas composition and temperature in 4×4 segments. Simulations were performed using a two-dimensional elementary kinetic model that represents the experimental setup in a detailed way. Excellent agreement between model and experiment was obtained for both global and local performance over all investigated operating conditions under varying H2/H2O/N2 compositions at the anode, O2/N2 compositions at the cathode, temperature, and fuel utilization. A strong variation of the electrochemical performance along the flow path was observed when the cell was operated at high fuel utilization. The simulations predict a considerable gradient of gas-phase concentrations along the fuel channel and through the thickness of the porous anode, while the gradients are lower at the cathode side. The anode dominates polarization losses. The cell may operate locally in critical operating conditions (low H2/H2O ratios, low local segment voltage) without notably affecting globally observed electrochemical behavior.

Keywords: Solid oxide fuel cell (SOFC), Segmented cell, Experiment, Modeling, Simulation

1 Introduction
High electrical efficiency and long-term durability are key requirements for a cost-competitive introduction of fuel cell technology. In order to achieve high efficiency, a high fuel utilization is required, that is, the complete fuel should be converted electrochemically in the fuel cell. In solid oxide fuel cells (SOFC), high fuel utilization results in strong concentration gradients at the anode, where the fuel (H2, CO, CH4) is successively diluted by reaction products (H2O, CO2). This, in turn, leads to an inhomogeneous distribution of electrochemical performance (local power density) and temperature. These inhomogeneities detrimentally affect both efficiency (through a large concentration polarization) and long-term stability (through thermomechanical stresses and local operating conditions that may induce degradation). In order to understand these effects, it is necessary to obtain detailed information about the spatial distribution of the electrical, chemical and thermal cell properties.
For polymer electrolyte fuel cells (PEFCs), segmented cell setups for the local measurement of current density have been developed and are widely used. In contrast to this, only few experiments for a spatially resolved characterization of SOFCs have been conducted so far, mainly because the high operating temperature (700-900 °C) represents a considerable challenge to the experimental setup. One of the first setups for the local characterization of segmented planar SOFC cells has been developed by Metzger et al. Segmented-cell setups are typical model experiments in the sense that they allow a largely extended insight into the cost of modified geometries and/or materials in comparison to standard fuel cells. Results from segmented cells are therefore not expected to be quantitatively equivalent to those from standard stacks in terms of performance or temperature distribution. Having this difference in mind, the accessible information is highly valuable both for increasing the fundamental understanding and for optimizing cell and flow field design. The use of mathematical modeling in combination with experimental investigations can significantly enhance the understanding of the spatially resolved electrochemical behavior of the cell. For example, today's experimental techniques do not allow the measurement of concentration and current distributions inside the porous electrodes. However, it is the electrochemically active zone inside the electrodes that governs electrochemical performance as well as degradation behavior. For this reason, modeling studies are increasingly used to aid understanding of the fuel cell processes and to optimize electrode and cell design. The combination of simulations with segmented-cell experiments is particularly useful because it allows reliable model validation including local behavior. Furthermore, the segmentation can easily be omitted and geometrical parameters be modified in the model, thus allowing to bridge the difference between the segmented cells and standard cells. In this paper we present a combined modeling and experimental study of the spatial distribution of electrochemical performance in a planar SOFC. The experiments are based on the setup by Metzger et al. The simulations are conducted within a physically-based modeling framework developed by Bessler et al., which is extended in order to reflect the segmentation of the experimental setup. The following approach is used: (i) The model is validated using experimental polarization data under specifically designed calibration conditions; (ii) model predictions are compared with experimental data at realistic operating conditions; and (iii) the validated model is used for quantitative interpretation and prediction of the physical, electrochemical and fluid mechanical fuel cell properties.

2 Experimental Setup

The measurement setup for segmented cells has been described previously. It is shown schematically in Fig. 1a. Square-shaped anode-supported cells (100 cm² area, 540 µm Ni/YSZ cermet anode with a thin anode functional layer, 5 µm YSZ electrolyte, 3 µm YDC interlayer, 45 µm LSCF cathode divided into 16 segments with an active area of 4.84 cm² of each segment) were integrated in a metallic cell housing and sealed with glass seal (Fraunhofer Institute of Ceramic Technologies and Sintered Materials (IKTS), Dresden, Germany). The metallic housing is subdivided into 16 galvanically isolated segments with a total active area of 73.96 cm². The flow field is identical for fuel and air side; it consists of square-shaped channels (2 × 2 mm, 5 channels per segment row) that are supplied via inlet and outlet manifolds that are symmetrical to each other (ca. 4 cm² per manifold). In order to determine the temperature at each segment, thermocouples are introduced in the metallic segments. Additionally, capillary tubes that correspond to the cathodic segments are integrated at the anode side at 16 measuring points to take samples of the anode gas to be analyzed by gas chromatography. The setup allows for the integral and spatially resolved measurement of current density and voltage; the local and integral determination of impedance data, the local measurement of temperature and the spatially resolved analysis of the fuel gas concentrations along the flow path. Experiments were conducted under a wide range of operating conditions as summarized in Table 1. Conditions for model calibration were defined that show a high sensitivity of simulated polarization curves on unknown model parameters. The calibration measurements were carried out under high gas flow rates (low fuel utilization) in order to avoid strong fuel depletion. Even for the highest fuel dilution (Table 1, condition 6), fuel utilization was < 26 %. Additional measurements were performed under realistic gas compositions at high fuel utilization (Table 1, condition 10).

3 Modeling and Simulation

3.1 Planar SOFC Modeling Framework

The framework for physically-based SOFC modeling and simulation has been presented previously. The model layout is shown schematically in Fig. 2, and all model parameters are summarized in Table 2. A 1D+1D elemental kinetic electrochemical model was applied that represents one single channel of the experimental setup (Fig. 2b). One-dimensional channel flow (x-dimension) is described using the Navier-Stokes conservation equations (continuity, species, momentum), corresponding essentially to a plug-flow model with axial diffusion. One-dimensional mass transport through the membrane-electrode assembly (MEA) (y-dimension) is described by coupled Fickian/Knudsen diffusion and Darcy flow. Charge transport in the solid
electrolyte and the electrolyte phase of the composite electrodes is described in two dimensions using Ohm’s law.

The results shown in this paper are calculated under the assumption of isothermal conditions. This assumption is justified because measured temperature variations along the flow path are relatively low (26 K at a current density of 1.4 A/cm²) due to rapid heat losses via the metallic flow field and housing.

Anode electrochemistry is described using elementary kinetics for surface reactions on Ni and YSZ, and for charge transfer (hydrogen spillover) (Fig. 2c), as given in Table 3, and as validated by previous cermet anode and patterned anode studies. The electrochemical double layer is described as simple capacitor. Cathode electrochemistry is described using a modified Butler-Volmer equation,

\[ i = \dot{i} \exp(-E_{\text{act}}/RT) \cdot \{ \exp(0.5F\eta/RT) - \exp(-0.5F\eta/RT) \}. \]  

A definition of all symbols and their units is given in Table 2. Current-voltage relationships are modeled by directly solving for the electric-potential distribution in the electrodes and electrolytes without using the Nernst equation. All models are implemented in the in-house numerical simulation software DENIS. For numerical simulation, the MEA was spatially discretized into 16 × 80 control volumes (x × y dimension). For identifying unknown model parameters, a Levenberg-Marquardt nonlinear least-squares fitting algorithm was used.

### 3.2 Segmentation

For the present study, the model described in Ref. was extended in order to include the cell segmentation of the experimental setup. The segmentation is represented by an electrical circuit model as shown in Fig. 2a. An arbitrary number and size of segments can be used at both anode and cathode. The segments are individually connected via switches S to outer power rails at anode and cathode side. These rails are assumed to be at constant potential \( \phi \) given by the outer load. The connection wires are assumed to be resistors \( R \), leading to a potential drop modeled via Ohm’s law,

\[ \Delta \phi = R \cdot i_{\text{seg}}, \]  

where \( R \) is the cell-area-specific wire resistance and the potential drop \( \Delta \phi \) depends on the segment current density \( i_{\text{seg}} \). Along the length of each individual segment the potential and therefore the local segment voltage \( U_{\text{local}} \) are assumed constant. The simulation input is the voltage \( U \) of the external load. The model yields the total current \( I \), the individual segment currents \( I_{i} \), as well as local segment voltages \( U_{\text{local}} \). The latter are determined experimentally via individual voltage probes (four-point measurements at each segment). The model can also be used to calculate global and local impedance \( Z \) and \( Z_{\text{local}} \), respectively.

For the present simulations, four segments are used at both anode and cathode, and all switches S are closed, representing the experimental setup. The wire resistance \( R \) is determined via fitting to the experiments (cf. Table 2).

### 4 Results and Discussion

#### 4.1 Calibration Measurements and Model Validation

There are a number of model parameters associated with electrochemistry and transport in the MEA that are specific for the investigated cells and cannot be obtained from literature. Some parameters can be reliably estimated (e.g., porosity from SEM micrographs). Other parameters, however, can only be obtained by fitting to experimental data. To this end, the model was validated under specifically designed operating conditions (Table 1, conditions 1-9). Experimental polarization behavior was measured under low fuel utilization (< 15 %) by using fast gas inflow velocities in order to circumvent spatial gradients along the channel. Gas-phase anode and cathode inlet concentrations as well as temperature were varied over a range of conditions; these conditions were chosen in order to achieve sensitivity for the unknown parameters. Parameters were fitted using a one-dimensional model of the MEA only. Fitting results are included in Table 2 and Table 3.

In the present study, the following model parameters were fitted: (1) The preexponential factor of the elementary kinetic hydrogen spillover reaction at the anode (all other elementary kinetic parameters were used without modification from previous cermet anode studies); (2) the exchange current density of the global kinetic charge-transfer reaction at the cathode; (3) the \( p_{\text{OH}} \) exponent for cathode; (4) the tortuosity of the anode; (5) the ohmic resistance of the current collector wires. Given the high level of physicochemical detail of the present model, the number of free fit parameter has to be considered very low. It should be noted that the preexponential factor of the anodic charge-transfer reaction had to be modified by only 30 % in comparison to previous cermet anode studies; this small modification can be interpreted as variations in material and/or microstructure and demonstrates how elementary kinetic approaches can reduce empiricism in fuel cell modeling.
Simulated and experimental polarization curves for the validation conditions are shown in Fig. 3. There is
good quantitative agreement between model and experiment over the complete range of operating conditions
(anode H₂/H₂O ratio, anode gas dilution with N₂, cathode O₂/N₂ ratio, and temperature). Note that the physical
model uses only one single set of parameters for all conditions. A systematic deviation between model and
experiment occurs during temperature variation. We believe that this is due to an onset of cell degradation,
lowering the experimentally observed performance relative to the model predictions. Degradation effects are not
included in the model.

4.2 Measurement and Simulation Under High Fuel Utilization

The segmented cell was operated under a condition with high fuel utilization (Table 1, condition 10). The
anode was fed with 33% H₂, 1% H₂O and 66% N₂. This condition was chosen in order to simulate nitrogen-rich
reformate gas. The cathode was fed with air in counter-flow operation.

The measured two-dimensional distribution of power density in the 16 segments is shown in Fig. 4 for an
average power density of 460 mW/cm². The fuel utilization at this condition is 80%. The cell performance is
strongly inhomogeneous, with the power density systematically decreasing from fuel inlet (left side of Fig. 4) to
fuel outlet (right side of Fig. 4). It will be shown below that this decrease is due to fuel depletion along the flow
path. Moreover, there is a notable difference for the four segment rows (upper row to lower row in Fig. 4). This
difference may be due to sealing issues or inhomogeneous gas supply to the gas channels. Further segment-to-
segment scattering is likely due to a variation in contact resistance, which may also lead to systematic row-to-
row variations when the contact pressure is inhomogeneous. For comparison with the model (which represents
the behavior along one single channel), the row with segments 9-12 was chosen.

Experimental and simulated global and local current-voltage characteristics for segments 9-12 are shown in
Fig. 5. Simulations were performed using the 2D model (i.e., along one representative channel and through the
thickness of the MEA, cf. Fig. 2). The global IV-curve (Fig. 5a) shows a typical shape with a parabolic behavior
at low currents, linear behavior at intermediate currents, and a limiting current density of ~0.8 A/cm² at high
currents. The maximum power density (P_max = 470 mW/cm²) is observed at a cell voltage of 0.70 V. Local IV-
curves for segments 9-12 are represented by plotting local segment voltage versus local segment current (Fig.
5b). The local behavior shows a considerable variation of current density for different cell segments. At high
polarization, segments 11 and 12 that are located close to the fuel outlet show a particularly interesting behavior:
the current decreases while at the same time the segment voltage strongly decreases. This "inverse" behavior is
due to strong fuel depletion (see next section). At the same time, the current density of segment 9 that is located
at the fuel inlet continues to increase.

There is excellent quantitative agreement between model and experiment for both the local and the global
behavior. Note that no additional parameter adjustment was performed after model calibration (Section 4.1). This
finding also underlines the validity of the choice of calibration conditions.

4.3 Distribution of Gas-Phase Concentrations

Fig. 6 shows the simulated spatially resolved distribution of gas-phase concentrations in the channels as well
as in the porous electrodes and current collector meshes for the condition of maximum power density (P_max in
Fig. 5a). There are considerable concentration gradients throughout the complete system. In particular, the
relatively thick porous anode in this anode-supported cell is responsible for strong H₂ depletion through the
anode thickness. This is the case, in particular, for segments 11 and 12. Here, H₂ concentration tends towards
zero at the porous electrode/solid electrolyte interface. This is the reason for the strong decrease in segment
voltage observed in the local IV-curves (Fig. 5b).

The upper panel of Fig. 6 also shows the local current density along the flow path. The step-like behavior of
the four segments is due to the contribution of the resistance of current collector wires (cf. Fig. 2a): The potential
drop over the wire for the individual segments decreases as the overall segment current decreases along the fuel
flow path (left to right in Fig. 6). This leads to an increase in integral segment current, causing a step from the
end of one segment to the beginning of the next segment. Note that the segment voltage is measured and
simulated directly at the segment (U_local in Fig. 2a), therefore including the wire resistance. The current density
distribution along the length of each segment is inhomogeneous. The integral current density over one segment is
shown in Fig. 5b.

The condition of strong fuel depletion observed inside the porous anode may be highly critical concerning
cell degradation. In particular, low H₂/H₂O ratios may lead to nickel oxidation at the anode, and low local
segment voltages may lead to cathode degradation. Note that the critical behavior observed here in particular
at segment 12 is not obvious from the global IV-behavior. Thus, a cell may be locally in a critical operating
condition while global characteristics are well within apparent stability limits (e.g., sufficiently high global cell
voltage). Post-test analysis and interpretation with a focus on degradation is subject of ongoing studies.

4.4 Distribution of Polarization Losses
The knowledge of the physical origin of polarization losses is a requirement for an optimization of cell design and operating conditions. The validated model offers the possibility to quantify the spatially varying polarization losses. Results for segments 9 and 12 are shown in Fig. 7 for the same operating conditions as shown in Fig. 5. The total polarization losses are separated into contributions from Ohmic resistance of the electrolyte \( \eta_{\text{ohm}} \), concentration polarization \( \eta_{\text{conc}} \), at both anode and cathode, and activation polarization \( \eta_{\text{act}} \) at both anode and cathode.

For both segments, concentration polarization at the anode is the dominant loss process. This finding is consistent with the observation of strong H\(_2\) depletion along the channel and through the anode thickness. The second largest contribution is anode activation. Polarization due to the electrolyte and cathode activation and concentration losses are low and do not significantly affect the cell performance. With increasing current density, all losses increase up to the point of maximum power density. Beyond that point, Ohmic and activation losses decrease in segment 12 due to the decreasing local current density in these segments (cf. Fig. 5b).

The analysis shows that the parabolic behavior at low current density that is observed in the global IV-curve (Fig. 5a) is due to anode concentration polarization and not, as commonly assumed, due to activation polarization. This has been pointed out before\(^\text{14}\). In this respect, SOFCs behave differently from PEFCs where low-current parabolic behavior is indeed due to activation losses.

## 5 Conclusions

A combined experimental and modeling study of the spatial distribution of the electrochemical performance in a planar SOFC was performed. Experimental data were obtained using a specifically designed segmented cell setup that allows for the measurement of local current-voltage characteristics, gas composition and temperature. Simulations were performed using a two-dimensional elementary kinetic model that represents the experimental setup in a highly detailed way.

Model parameters were identified by comparing simulations with validation experiments under low fuel utilization. Experiments and simulations under high fuel utilization showed strong gradients of gas concentrations and current density along the flow path and throughout the thickness of the membrane-electrode assembly. Over the complete range of operating conditions, excellent quantitative agreement between model and experiment for both global and local behavior was observed. The relatively thick porous anode and high fuel utilization causes particularly strong concentration variations at the anode, while the gradients are lower at the cathode side. The combined modeling and experimental study allows for the reliable quantification of the contribution of various polarization losses. It was shown that concentration polarization at the anode is the dominant loss process, followed by anode activation polarization. The anode was therefore identified as the critical component in this cell type.

An important conclusion is that the cell may operate locally in critical operating conditions (low H\(_2\)/H\(_2\)O ratios, low local segment voltage) without notably affecting global electrochemical behavior. These conditions are likely to detrimentally affect cell durability. Based on a combination of experiment and modeling, the present study allows for identifying such conditions and for assessing optimized cell design and operation strategies.

## Acknowledgements

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## References


Figure Captions

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Fig. 4: Measured two-dimensional distribution of power density over the 16 segments under operating conditions with high fuel utilization (counter-flow operation; anode: 33 % H₂, 1 % H₂O, 66 % N₂ in H₂, 1.1 m/s inflow velocity; cathode: air, 5.2 m/s inflow velocity; T = 800 °C) at a cell voltage of 0.59 V. The fuel inlet is at the left side, the air inlet at the right side.

Fig. 5: Experimental and simulated polarization behavior for the segmented cell using the 2D model under operating conditions with high fuel utilization (counter-flow operation; anode: 33 % H₂, 1 % H₂O, 66 % N₂ in H₂, 1.1 m/s inflow velocity; cathode: air, 5.2 m/s inflow velocity; T = 800 °C). (a) Global current-voltage curves, (b) Local segment voltage versus local segment current. The numbers indicate the segments, where segment 9 is the first in flow direction of the fuel gas.

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Tables

<table>
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<th>% H₂O</th>
<th>% N₂</th>
<th>Cathode % O₂</th>
<th>% N₂</th>
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Table 1: Experimental measurement conditions. Conditions 1-9 represent calibration conditions (low fuel utilization and specific N₂ dilution), condition 10 a realistic operating condition (high fuel utilization).

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<tr>
<td><strong>Porous media (mesh anode (ma), cermet anode (an), porous cathode (ca), mesh cathode (mc))</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td><strong>Cermet anode</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Active three-phase boundary length</td>
<td>4.5 · 10⁻¹² m³</td>
<td>Micro-structural model¹⁵</td>
</tr>
<tr>
<td>Effective width electrolyte</td>
<td>0.43 µm</td>
<td>Setup</td>
</tr>
<tr>
<td>Effective width electrode</td>
<td>0.31 µm</td>
<td>Setup</td>
</tr>
<tr>
<td>Structural factor electrolyte conductivity</td>
<td>0.30</td>
<td>Setup</td>
</tr>
<tr>
<td>Electrolyte thickness</td>
<td>10 µm</td>
<td>Setup</td>
</tr>
<tr>
<td>Conductivity bulk 8YSZ: preexponential factor, activation energy</td>
<td>5.15·10⁷ SK/m, 84 kJ/mol</td>
<td>Setup</td>
</tr>
<tr>
<td>Additional resistances</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Resistance of wires</td>
<td>1.4 Ωcm¹⁴</td>
<td>Exp.</td>
</tr>
<tr>
<td><strong>Conditions</strong></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Temperature</td>
<td>700 - 800 °C</td>
<td>Exp.</td>
</tr>
<tr>
<td>Pressure</td>
<td>1013 hPa</td>
<td>Exp.</td>
</tr>
</tbody>
</table>

¹ Estimated.
² Fitted to the present experiments.

Table 2: Model parameters used for all calculations.
Anode reaction

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Preexponential factor</th>
<th>Activation energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>Charge transfer reaction (Fit):</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\text{Ni}<em>\text{e} + \text{OH}</em>\text{YSZ} \rightleftharpoons \text{Ni} + \text{H}<em>2\text{O}</em>\text{YSZ} + e_\text{Ni}$</td>
<td>$1.63 \times 10^3 \text{ mol/(m s)}^{a}$</td>
<td>$235 \text{ kJ/mol}^{b}$</td>
</tr>
<tr>
<td>YSZ surface reactions:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\text{H}<em>2\text{O} + \square</em>{\text{YSZ}} \rightleftharpoons \text{H}<em>2\text{O}</em>{\text{YSZ}}$</td>
<td>$6.6 \times 10^{11} \text{ mol/(cm}^3 \text{s})$</td>
<td>$0$</td>
</tr>
<tr>
<td>$\text{H}<em>2\text{O}</em>{\text{YSZ}} + \text{O}^{2-}<em>{\text{YSZ}} \rightleftharpoons 2 \text{OH}</em>{\text{YSZ}}$</td>
<td>$1.6 \times 10^{22} \text{ mol/(cm}^3 \text{s})$</td>
<td>$9.6 \text{ kJ/mol}$</td>
</tr>
<tr>
<td>Nickel surface reactions:</td>
<td></td>
<td></td>
</tr>
<tr>
<td>$\text{H}<em>2 + \square</em>{\text{Ni}} \rightleftharpoons 2 \text{H}_{\text{Ni}}$</td>
<td>$9.8 \times 10^{17} \text{ mol/(cm}^3 \text{s})$</td>
<td>$0$</td>
</tr>
<tr>
<td>$\text{H}<em>2\text{O} + \square</em>{\text{Ni}} \rightleftharpoons \text{H}<em>2\text{O}</em>{\text{Ni}}$</td>
<td>$1.4 \times 10^{10} \text{ mol/(cm}^3 \text{s})$</td>
<td>$0$</td>
</tr>
<tr>
<td>$\text{H}<em>2\text{O}</em>{\text{Ni}} + \text{O}<em>{\text{Ni}} \rightleftharpoons \text{OH}</em>{\text{Ni}} + \square_{\text{Ni}}$</td>
<td>$5.0 \times 10^{27} \text{ mol/(cm}^3 \text{s})$</td>
<td>$98 \text{ kJ/mol}$</td>
</tr>
<tr>
<td>$\text{H}<em>2\text{O}</em>{\text{Ni}} + \text{O}<em>{\text{Ni}} \rightleftharpoons 2 \text{OH}</em>{\text{Ni}}$</td>
<td>$5.4 \times 10^{21} \text{ mol/(cm}^3 \text{s})$</td>
<td>$209 \text{ kJ/mol}$</td>
</tr>
<tr>
<td>$\text{H}<em>2\text{O}</em>{\text{Ni}} + \text{OH}<em>{\text{Ni}} \rightleftharpoons \text{H}<em>2\text{O}</em>{\text{Ni}} + \square</em>{\text{Ni}}$</td>
<td>$3.0 \times 10^{29} \text{ mol/(cm}^3 \text{s})$</td>
<td>$43 \text{ kJ/mol}$</td>
</tr>
</tbody>
</table>

Cathode reaction

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Exchange current density</th>
<th>Activation energy</th>
<th>Exponent $\alpha$ for $p_{\text{O}_2}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\frac{1}{2} \text{O}<em>2 + \text{V}^{**}</em>{\text{YSZ}} + 2 e \rightleftharpoons \text{O}^{x}_{\text{YSZ}}$</td>
<td>$4.4 \times 10^{10} \text{ A/m}^2$</td>
<td>$125 \text{ kJ/mol}$</td>
<td>$0.47$</td>
</tr>
</tbody>
</table>

$^{a}$ Fitted to the present experiments.
$^{b}$ Fitted to experiments with symmetrical SOFC cermet anodes.
$^{c}$ Activation energy from dense thin film electrodes.

Table 3: Elementary kinetic reaction mechanism for the anode and global kinetic reaction for the cathode. A single hydrogen spillover between Ni and YSZ surfaces is assumed as active charge-transfer reaction with a symmetry factor $\alpha = 0.5$. The surface site densities of Ni and YSZ are $6.1 \times 10^{-9}$ and $1.3 \times 10^{-8} \text{ mol/cm}^2$, respectively.