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ENHANCING THE LIFETIME OF SOFC STACKS FOR COMBINED HEAT AND POWER APPLICATIONS

SOF-CH

Final Report ZHAW-ICP 2007: **WP 5.1**: THERMO-MECHANICS

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RÉSUMÉ

The Finite Element (FE) simulations performed at the Institute of Computational Physics (*ICP*) of the Zurich University of Applied Sciences in Winterthur (*ZHAW*) aim at investigating mechanical stresses, induced into the various components of the *Hexis* SOFC-stack under different operation conditions. In general, internal stresses are caused by external forces, by mismatch of the thermal expansion coefficients of different stack components as well as by a non-homogeneous distribution of the stack-temperature. Once a critical stress level is reached, this can cause cell fracture which almost certainly leads to the complete failure of the affected cell.

In close collaboration with *Hexis*, *Empa-HLK* (Jakob Kübler), *Empa-MMS* (Prof. Eduoardo Mazza), and *NM Numerical Modelling GmbH*, simulations tools, using 2D and 3D rotational-symmetric FE-models, have been developed to describe the thermo-mechanical issues that occur in the stack. At first, the thermo-mechanical properties of all the stack components were extracted from measurements conducted by *Empa-HLK*. Temperature dependent Young's modulus and CTE-data where thus obtained.

The thermo-mechanical FE-analysis of the whole *Hexis*-stack and its mounting showed that the uniformity of the stresses throughout the stack strongly depends on the mounting and the MIC design. By using model-based optimizations, new concepts have been developed to improve the mounting and the design.

Buts du projet

The Finite Element (FE) simulations performed at the Institute of Computational Physics (*ICP*) of the Zurich University of Applied Sciences in Winterthur (*ZHAW*) aim at investigating mechanical stresses, induced into the various components of the *Hexis* SOFC-stack under different operation conditions. In general, internal stresses are caused by external forces, by mismatch of the thermal expansion coefficients of different stack components as well as by a non-homogeneous distribution of the stack-temperature. Once a critical stress level is reached, this can cause cell fracture which almost certainly leads to the complete failure of the affected cell.

Travaux effectués et résultats acquis

Employed Models

For the thermo-mechanical stress-analysis of the *Hexis* SOFC-stack, the following 3D and 2D rotational-symmetric FE-models have been developed and used:

1) **3D Thermo-mechanical-flow FE-model of a repeat-unit of the Hexis stack**
This model is used to investigate how non-homogeneous stack temperatures cause locally induced mechanical stresses in different cell-components. Since in the model, the main characteristics of the metallic interconnect (MIC) geometry have been parameterized, this tool is ideal for MIC-design optimizations.

2) **2D rotational-sym. mechanical FE-model of a cell mounted in a ring-ring device**
This nonlinear-elastic mechanical model is used to obtain temperature-dependent Young's modulus of whole cells from fitting force-versus-deformation curves measured at *Empa-HLK*.

3) **2D rotational-symmetric thermo-mechanical FE-model of a cell**
This linear-elastic thermo-mechanical model is used to calculate cells stresses and cell deflections for given temperatures and different mountings: the cell is either assumed to be clamped between two parallel plates, or it is allowed to freely deform. This model is used, for example, to obtain Young's modulus of individual cell-layers from Young's modulus of whole cells in combination with experimental deflection data. It is also used to verify measured thermal expansion coefficients (CTEs) by comparing the predicted cell deflections with experimental data.

4) **3D thermo-mechanical FE-model of a cell**
This linear-elastic thermo-mechanical model is used to investigate the effect of cracks in the anode- and cathode-layers on electrolyte stresses. This is relevant for cells, where the electrolyte is the supporting element (so-called electrolyte-supported cells). It has also been used to predict the 3D-shapes of freely deformable cells.

5) **3D mechanical FE-model of a repeat-unit (cell + interconnect)**
This linear-elastic mechanical model is used to calculate spatially averaged “effective” mechanical properties of a repeat-unit, which are subsequently used to characterize the mechanical behavior of whole stacks.

6) **2D rotational-symmetric thermo-mechanical FE-model of a stack**
This linear-elastic thermo-mechanical model is used to predict the stresses exerted on the stack by its external mounting.

For all calculations, our in-house multi-physics FE-code *Seses* (www.icp.zhaw.ch/seses) was used.

State at Intermediate Status Report on December 2006

Back in December 2006, the following outlook was given:

1) More accurate material properties measured at *Empa-HLK* are needed. The simulation results obtained so far are still partly based on literature values for different material properties, which are not considered reliable enough.

2) The procedure to get Young's modulus values of the cell components through nonlinear elastic FE-analysis needs to be refined. The results obtained so far show rather large variations.

3) The rotational-symmetric stack model based on the volume averaging approach needs to be used to optimize the stack mounting as well as the interconnect design with respect to the stress distribution.

4) The 3D thermal flow model needs to be tested against measured temperature distributions. It is also planned to include linear-elastic stress analysis on the cell level into this model. This will allow for optimizations of the stack design to reduce the stress levels.

5) The exertion of nonhomogeneous stresses on cell materials by partially oxidized, deformed interconnects has not yet been clarified. This includes realistic contact conditions between the interconnects and the cells.

The above points have been all addressed together with several new issues, that were not apparent back in 2006. Each topic will be briefly discussed in the following section.

State of Current Work

1. Determination of thermo-mechanical properties of cell materials

It has been a major challenge to accurately determine temperature-dependent Young's modulus data as well as thermal-expansion coefficients (CTEs) for all cells components, i.e. for the anode- and cathode-layers as well as for the electrolyte material. This is because the very brittle and fragile structure of SOFCs cause a large variability and large uncertainties in the experimental characterization of their thermo-mechanical properties. As an example, Figure 1 shows Young's modulus at two different temperatures as obtained from performing nonlinear-elastic FE-analysis of force-versus-deformation curves measured at *Empa-HLK*.

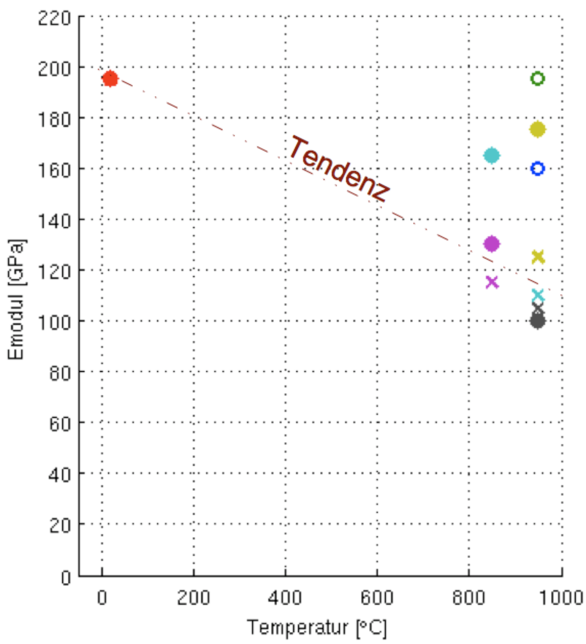

Figure 1: Young's modulus at different temperatures as obtained from applying nonlinear-elastic FE-analysis to force-versus-deformation curves measured at *Empa-HLK.*

Clearly, the data at high temperatures shows huge variations. Similar variations have been obtained from CTE-measurements of different electrode-layers. Fortunately, for both Young's modulus and CTE-data, plausibility checks can be made.

Cell layers	T (°C)	d_{sim} (mm)	d_{exp} (mm)	σ_{max} (MPa)	σ_{elec} (MPa)
E+A_1	20	3.30	3.33	103 (A_1)	30
E+A_1+A_2	20	7.00	6.70	90 (A_1)	60
E+K_1	20	0.85	0.80	26 (K_1)	5
E+K_1+K_2	20	1.20	1.20	24 (E)	24
Cell	20	3.86	3.90	100 (A_1)	25
Cell	20	0.00	0.00	116 (A_1)	25
Cell	950	0.00	0.00	0	0
Cell	930–950	0.00	0.00	26 (E)	26

Figure 2: Comparison of simulated deflections (d_{sim}) of freely deformable cells with experimental data (d_{exp}). Also shown are the corresponding mechanical stresses induced in different cell-layers.

For example, the obtained material parameters can be checked by using them in an FE-model to predict the deflections of freely deformable cells at different temperatures and by comparing the simulation results with experimental data. Such comparisons are shown in Figure 2. One sees that the calculated and measured deflections agree well with each other. Note also that the whole cell shows a deflection of 4 mm at room temperature.

2. External mounting and force-distribution in the *Hexis* SOFC-stack

As mentioned in the beginning, besides thermally induced stresses as caused by CTE-mismatch and local temperature-gradients, external forces induced by the stack mounting play an important role. In addition, these external forces have a strong influence on the sealing tightness of the SOFC-stack. The idea is to perform model-based optimizations of the stack-mounting and the interconnect-design to ensure a homogenous force-distribution as well as a gas-tight system. Figure 3 shows the method by which effective material properties of a repeat-unit are obtained from volume averaging.

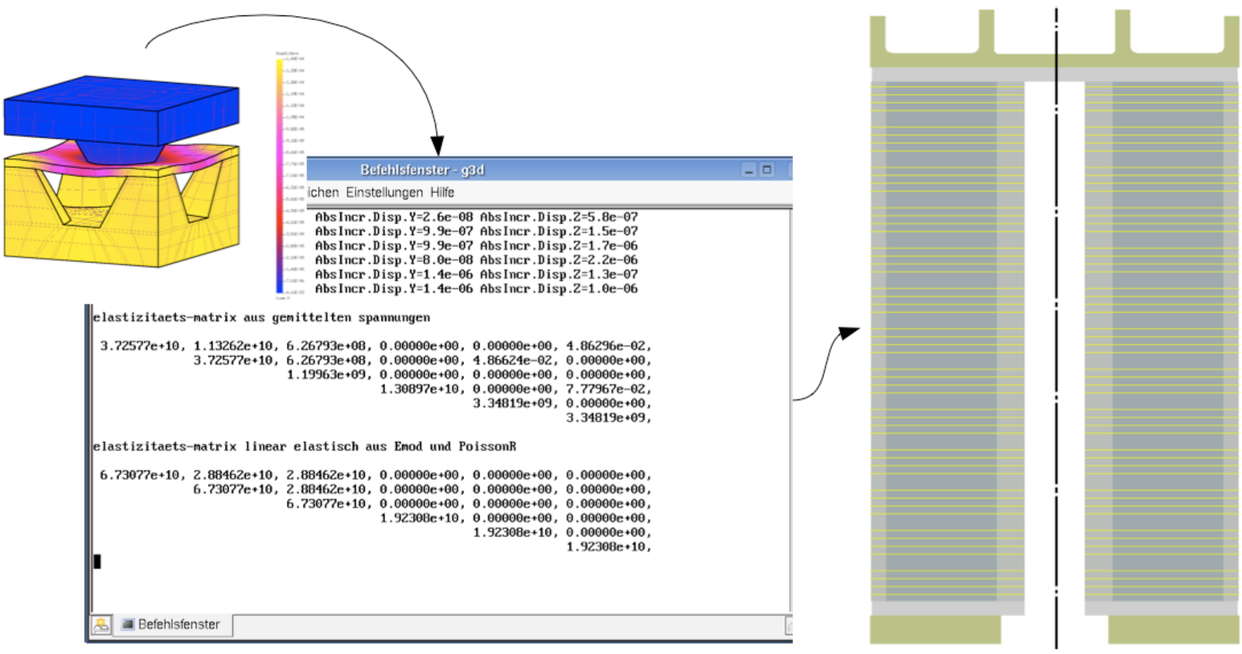

Figure 3: Averaging method to obtain effective mechanical properties of the stack. The idea is to determine spatially averaged mechanical properties from the stack-element, shown on the left. Once the effective mechanical properties are known, they can be used to calculate stresses and deformations of the stack in the rotational-symmetric configuration shown on the right.

Once the effective mechanical stack properties are known, they can be used to calculate the stresses and deformations of the whole stack in a simple rotational-symmetric configuration. This approach is highly efficient in terms of required RAM and CPU-time and is therefore ideal for extensive parameter studies – to optimize, for example, the external stack mounting with respect to a homogeneous stress-distribution in the stack. As an example, Figure 4 shows the distribution of axial stresses near the top of the *Hexis* SOFC-stack. (Compressive forces are indicated in green and blue, tensile forces in red. Stress-free zones are shown in black.) One sees that this specific mounting-design (in combination with the particular interconnect-design) causes a rather uneven force-distribution which might be a source for gas-leakages, especially at the stack perimeter, where compressive forces are low. To solve this problem, the model has been used to compare different designs for the stack-mounting with each other.

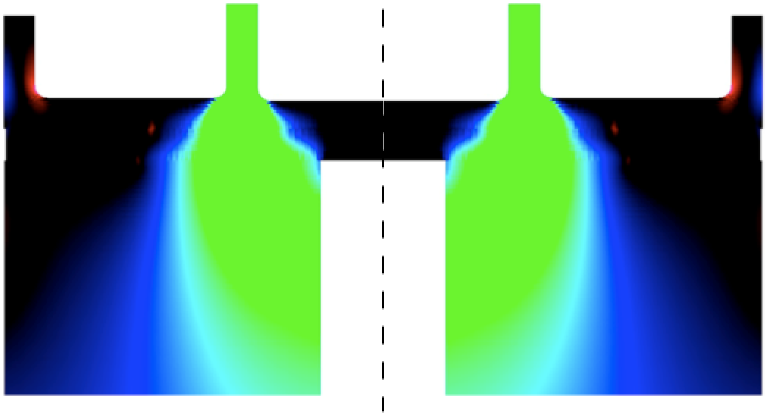

Figure 4: Axial stress-distribution near the top of the *Hexis* SOFC-stack. Compressive forces are indicated in green and blue, tensile forces in red. Stress-free zones are shown in black.

3. Thermo-mechanical stresses induced by local temperature-gradients

A 3D thermo-mechanical-flow FE-model of a repeat-unit of the *Hexis* stack has been developed. As shown in Figure 5, for symmetry reasons, only 1/8 of the repeat-unit needs to be taken into account. This model is used to investigate how non-homogeneous stack temperatures cause locally induced mechanical stresses in different cell-components. Since in the model, the main characteristics of the metallic interconnect (MIC) geometry have been parameterized, it is ideal for MIC-design optimizations.

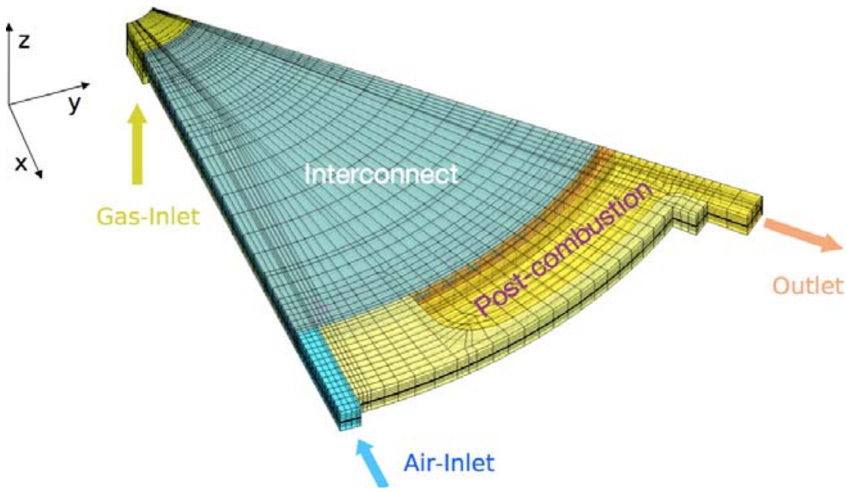

Figure 5: 3D thermo-mechanical-flow-model to obtain the flow and temperature distribution within a single repeat-unit and to calculate the stresses caused by local temperature-gradients.

The model comprises the cell components as well as the interconnect gas-channels on the anode- and cathode-sides. It also includes the post-combustion zone which is a major heat source. Note that the electrochemical and bulk-phase reactions taking place on the cells are not explicitly modeled.

However, the heat released on the cells as well as the heat consumption by internal reforming are obtained from another FE-model that includes all major chemical reactions and multi-component species transport. Figure 6 shows the temperatures of a cell mounted in a repeat-unit of the *Hexis* SOFC-stack for two different operation conditions: the results on the left are for standby OCV-conditions and the ones on the right for full-load-conditions. In OCV-mode, all heat is supplied from the post-combustion zones located at the perimeter of the stack. Consequently, the coolest temperatures occur in the center (where the fuel entry is located) and at the air-inlet channel (at the perimeter). In contrast, under full-load operation, much of the heat is released on the cells close to the fuel gas inlet, shifting the maximum temperature from the outside to the inside of the stack.

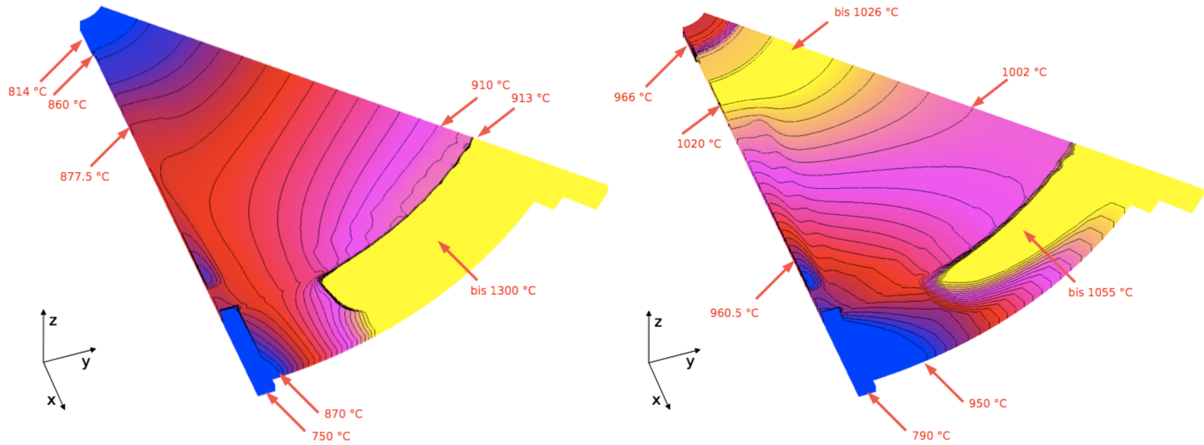

Figure 6: Temperatures of a cell mounted in a repeat-unit of the Hexis SOFC-stack under OCV-conditions (left) and full-load-conditions (right). The highest temperatures are indicated in yellow, the lowest in blue.

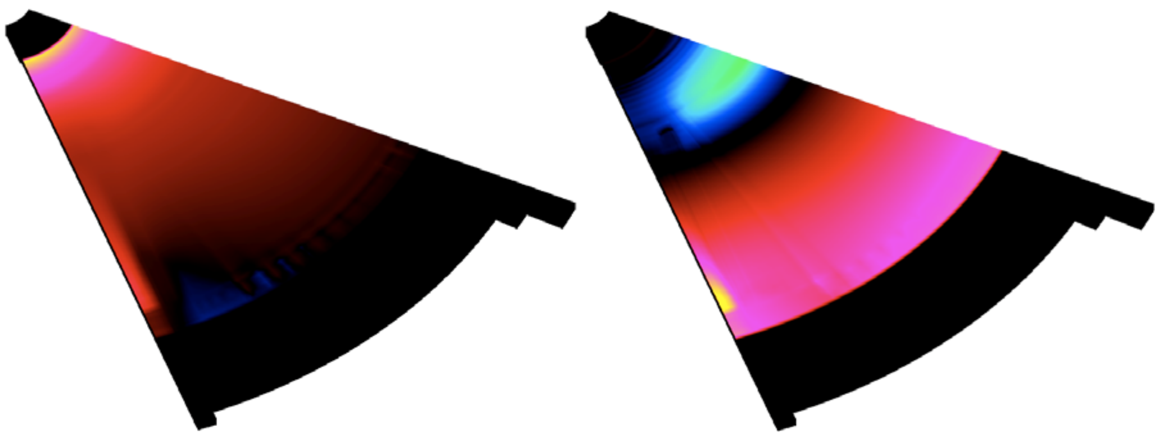

Figure 7: Maximum principal stresses of a cell mounted in a repeat-unit of the Hexis SOFC-stack under OCV-conditions (left) and full-load-conditions (right). The highest stresses are indicated in yellow, stress-free zones are shown in black and compressive stresses in blue and green.

Figure 7 shows the maximum principal stresses as induced by the temperature-profiles presented in Figure 6. (The highest stresses are indicated in yellow, stress-free zones are shown in black and compressive stresses in blue and green.) Under OCV-conditions, the highest tensile stresses occur in the center near the fuel inlet, whereas in full-load mode, the highest tensile loads occur near the air inlet at the stack perimeter. These results clearly indicate the importance of the heat-exchanger and insulation components surrounding the stack. When the temperatures of the gas-streams entering the stack deviate from the average stack temperature too much, this is a major source for thermo-mechanical stresses exerted to different cell components.

4. Influence of cracks on the mechanical integrity of the electrolyte material

In electrolyte-supported SOFCs such as the one used in the *Hexis*-system, cracks in the electrodes might be an important mechanism by which the electrolyte material is weakened. Such cracks occur during manufacturing, handling and operation. A 3D thermo-mechanical FE-model has been developed to analyze this effect. As shown in Figure 8, the model contains two artificially introduced cracks in the radial and azimuthal directions. The picture to the right shows a typical stress-distribution in the vicinity of a crack. One sees that the highest tensile stresses (indicated in yellow) occur right at the electrode-electrolyte interface. This clearly demonstrates the crack-induced weakening effect the electrodes can have on the electrolyte.

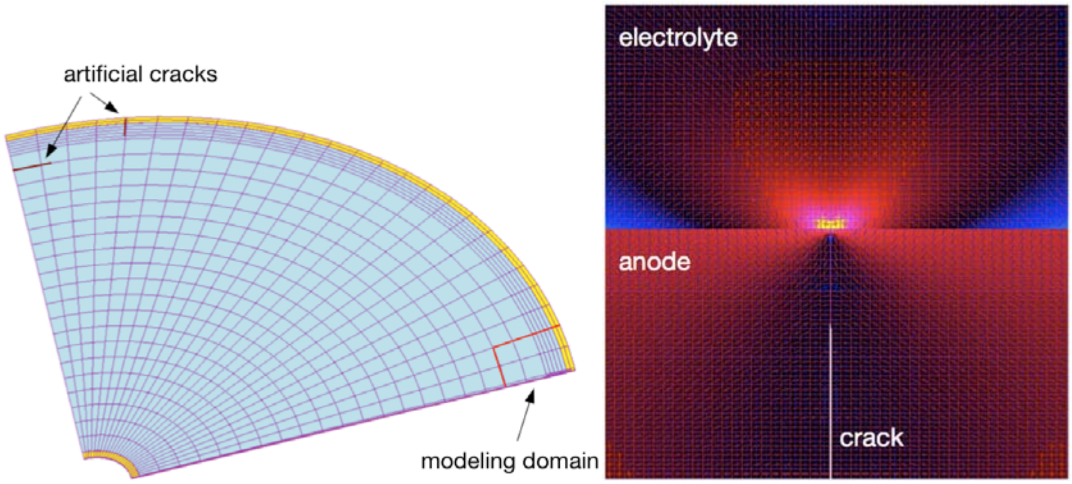

Figure 8: 3D FE-model of a cell with artificially introduced cracks in the radial and azimuthal directions. The FE-grid is shown on the left and the picture on the right shows the calculated stresses around a crack near the interface between the electrode and the electrolyte.

In a further step, the influence of different crack-populations on the cell stresses and cell deflections has been analyzed. Figure 9 shows the typical deflection of a newly manufactured cell at room temperature. The predicted shape agrees well with experimental observations. This is an impressive example of how the employed models can be used to identify microscopic phenomena that that can hardly be understood solely by experimental methods.

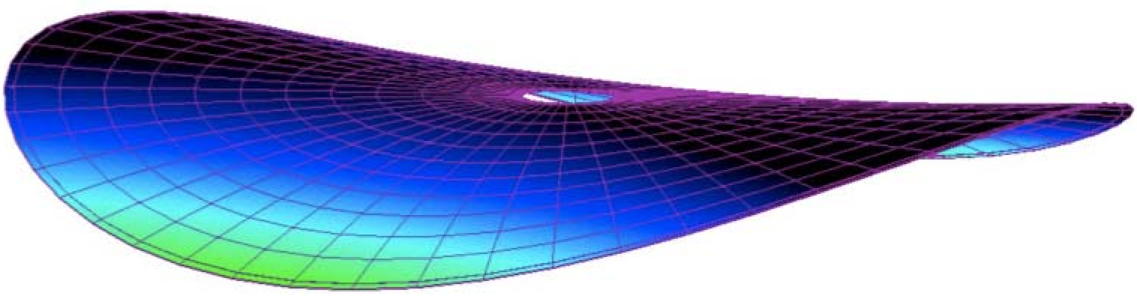

Figure 9: Prediction of the deflection-behavior of a cell as a combination of its thermo-mechanical material properties and a certain distribution of electrode-cracks.

Collaboration nationale

In 2006, *Hexis*, *Empa-HLK* and *Empa-MMS* and *ZHAW-ICP* (with support from *NM Numerical Modelling* GmbH, Thalwil) started their joined efforts to investigate the thermo-mechanical behavior of the *Hexis* SOFC-stack. The close collaboration between system developers, modeling experts, and material scientists was crucial and gave substantial insight into the different phenomena by which cell-breakage occurs.

Évaluation de l'année 2007 et perspectives

Based on this understanding, several new concepts have been developed and further refined by model-based parameter optimizations. They show promise to significantly lower the levels of thermo-mechanical stresses induced in the cells. Specifically, the following results have been obtained:

1. Temperature-dependent Young's modulus for the electrolyte and the different electrode-layers have been obtained from correlating the FE-model for the ring-ring setup with force-versus-deformation curves measured at *Empa-HLK*. The obtained values at high temperatures show huge variations. However, these variations could be substantially reduced by checking its consistency and plausibility when used to predict cell-deflections at different temperatures and comparing them with experimental data. Still, it would be desirable to get more accurate Young's modulus data at high temperatures – especially for the electrolyte, which is the supporting cell-layer in the *Hexis* cell.

2. The situation is similar concerning the determination of temperature-dependent CTE-data. Here as well, the data – especially for the anodes – showed huge variations, however this time at lower temperatures. By making the above mentioned plausibility checks and by comparing the obtained results with literature data (e.g. F. Tietz, *Ionics*, vol. 5, pp. 129 – 139, 1999 and F. Tietz et al., *Solid State Ionics*, vol. 177, pp. 1753 – 1756, 2006) the variations could be reduced. Still, it would be desirable to get more accurate CTE-data especially for the anodes, which are mainly responsible for the residual stresses induced in the electrolyte.

3. The low-temperature Weibull-data determined at *Empa-HLK* provides statistically sound information about the stress-levels that can cause cell fracture. (To obtain information about stress-levels that are critical at high temperatures, the low-temperature data was extrapolated based on literature information and experience by our partners at *Empa-HLK* and *Empa-MMS*.) It became obvious that both stresses caused by CTE-mismatch of adjacent layers as well as those caused by local temperature-gradients are within the critical range. Clearly, improvements are necessary both on the cell- and the system-design. To perform the required parameter-optimizations, the FE-models mentioned in the "employed models" section have proven to be powerful. However, the development of new concepts for the cell- and system-design is still ongoing and will probably extend to midyear of 2008. To further improve on the available pool of material data, it would be useful to have Weibull-statistics also available at high temperatures.

4. The thermo-mechanical FE-analysis of the whole *Hexis*-stack and its mounting showed that the uniformity of the stresses throughout the stack strongly depends on the mounting- and the MIC-design. (A uniform distribution of compressive stresses throughout the stack is critical for good contacting of the different stack-layers as well as for minimal leakage-losses, especially at the inner and outer stack-boundaries.) By using model-based optimizations, new concepts have been developed to improve the mounting- and the MIC-design.

Furthermore, as has become obvious during the last months, certain thermo-mechanical phenomena are tightly linked with the manufacturing process, the system dynamics and operations modes such as redox- and thermo-cycles. The latter leads to various degradation phenomena which in turn influence the mechanical integrity of the cells. Therefore, we plan to carry out further thermo-mechanical investigations within the *SOF-CH*-project as follows:

1. Thermo-mechanical analysis of the manufacturing process of the cells.

2. Continued thermo-mechanical analysis of different operations conditions including the system dynamics

3. Continued optimization of the MIC-design and the stack-mounting to reach a more homogeneous stress distribution and to reduce gas-leakages.

4. Extend thermo-mechanical models to include the impact of degradation phenomena such as redox- and thermo-cycles.

Annexes

Financial report.

ZHAW-project no. 57051.5.107.01 (“old” project no.)

Expenditures: CHF 17’580

57051.5.107.01 Hexis SOFC

Projekt	IST				BUDGET				Abweichung zu Budget			
Leistungstyp/-art	Stunden	Kosten	Ertrag	Erfolg	Stunden	Kosten	Ertrag	Erfolg	Stunden	Kosten	Ertrag	Erfolg
2059. Hocker Thomas	81	-10'530	0	-10'530	0	0	0	0	-81	-10'530	0	-10'530
4780. Tiefenauer Andreas	118	-7'050	0	-7'050	0	0	0	0	-118	-7'050	0	-7'050
	199	**-17'580**	**0**	**-17'580**	**0**	**0**	**0**	**0**	**-199**	**-17'580**	**0**	**-17'580**