PROJECT FINAL REPORT

"**Publishable**"

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1 Executive summary

On 1^{st} of May 2013, the research project PROSOFC started under the coordination of AVL List GmbH (Austria). It was the goal of PROSOFC to significantly improve the stack robustness and cost with the focus on production and operation. The first period was based on the Topsoe stack design. Due to the termination of Topsoe's SOFC activities by the end of August 2014, SOLIDpower took over the role of the industrial stack supplier to continue the project by keeping the main targets.

PROSOFC directly contributes to the objectives of Call SP1-JTI-FCH.2012.3.2 "Improved cell and stack design and manufacturability for application specific requirements".

- Improved electrical efficiency over the state of the art
- Better robustness, including longer lifetime
    - Operation in simulated real-life environment > 4,000 hours
    - Demonstration of the potential to achieve longer run times required to meet market entry requirements
- Considerable cost reductions consistent with market acceptance requirements for industrial or residential or other relevant applications
- Improved manufacturing methods in terms of yield and cost, reducing stack scrap rate to 10% by 2014 and the objective to reduce it to less than 5% by 2017
- Higher power density

In particular the PROSOFC project aims at improving the robustness, manufacturability, efficiency and cost of SOLIDpowers state-of-the-art SOFC stacks so as to reach market entry requirements. The key issues are the mechanical robustness of solid oxide fuel cells (SOFCs), and the delicate interplay between cell properties, stack design, and operating conditions of the SOFC stack.

The novelty of the project lies in combining state of the art methodologies for cost-optimal reliability-based design (COPRD) with actual production optimization. To achieve the COPRD beyond state of the art multi-physical modeling concepts had to be developed and validated for significantly improved understanding of the production and operation of SOFC stacks. The models should allow a probabilistic approach to consider statistical variations in production, material and operating parameters for the optimization phase. The key to this understanding are validating experiments and models on multiple levels of the SOFC system and introduction of extensive test programs specified by the COPRD methodology. In this context, an accelerated test program was developed which specifically covers the stresses coming from the operation of a stationary system (heat-up, cool-down, hot-standby, load changes, etc.).

2 Project context and objectives

Within the project an innovative methodology for a production and reliability oriented SOFC cell and stack design was developed. In particular the PROSOFC project aimed at improving the robustness, manufacturability, efficiency and cost of SOLIDpower state-of-the-art SOFC stacks so as to reach market entry requirements. The key issues are the mechanical robustness of SOFCs, and the delicate interplay between cell properties, stack design, and operating conditions of the SOFC stack.

The novelty of the project lies in combining state of the art methodologies for cost-optimal reliability-based design (COPRD) with actual production optimization. To achieve the COPRD beyond state of the art, multi-physical modeling concepts were developed and validated for significantly improved understanding of the production and operation of SOFC stacks. The most critical failure modes for stack operation were identified in order to align the experimental investigations to generate suitable data for modelling these failures. The multi-physics model allows a probabilistic approach to consider statistical variations in production, material and operating parameters for the optimization phase. The project provides a methodology for 3D description of spatial distribution of material properties based on a random field models. The key for the whole methodology are validating experiments, homogenized models on multiple levels of the SOFC system (cell, interconnect, sealings) and introduction of extensive test programs specified by the COPRD methodology. The probabilistic models were related to the experimentally obtained properties of base materials to establish a statistical relationship between the material properties and the most relevant load effects with respect to potential damage/failure modes. Software algorithms for meta models that allow the detection of relationships between input parameters (e.g. materials, loading, etc.) and output parameters (e.g. temperatures, stress, etc.) and to perform a sensitivity analysis were developed and implemented. The capabilities of the methodology for COPRD being developed was illustrated on several practical cases.

Following objectives were defined for the project

- Identification of the most important failure modes inside the stacks which should be covered by the modeling approach
- Comprehensive material (cell, interconnect, sealing) characterization to develop homogenized models to describe the material properties used in multi-physics stack modelling
- Comprehensive electrochemical cell characterization to develop homogenized models to describe the cell behavior used in multi-physics stack modelling
- Provide a methodology for 3D description of spatial distribution of material properties based on a random field model
- Establish a statistical relationship between the material properties and the most relevant load effects in view of potential damage/failure based on multi-physics simulation
- Provide both a physical model based as well as a purely statistically based description of the production process including the effect of non-destructive testing and screening
- Establish computationally efficient multi-physics meta-models of stack conditioning and operation to be utilized for automatic design variation and cost-based design optimization
- Carry out an automated sensitivity analysis with the software tool optiSlang in combination with the design software tools (e.g. Fluent, gProms, AVL FIRE, matlab, Simulink)
- Development of stack test program and calculation of demonstrable reliability
- Perform long term stack testing to validate stack design optimization
- Use of post mortem analysis to support stack robustness validation
- Disseminate PROSOFC methodology through several workshops and conference sessions, scientific papers and a webpage

3 Main scientific and technical results and foregrounds

3.1 Complex mechanical continuum material laws for stack modelling

To predict possible failure with the stack models in WP4, the mechanical material parameters must be known. It is shown in ref. 1 that creep influence the stress in the SOFC stacks over time and thus influence the probability of failure of the cells, see Figure 1. Here the probability of failure is seen to peak, when the operational conditions are changed. Between shifts in operation the stress relaxes governed by the new temperature distribution.

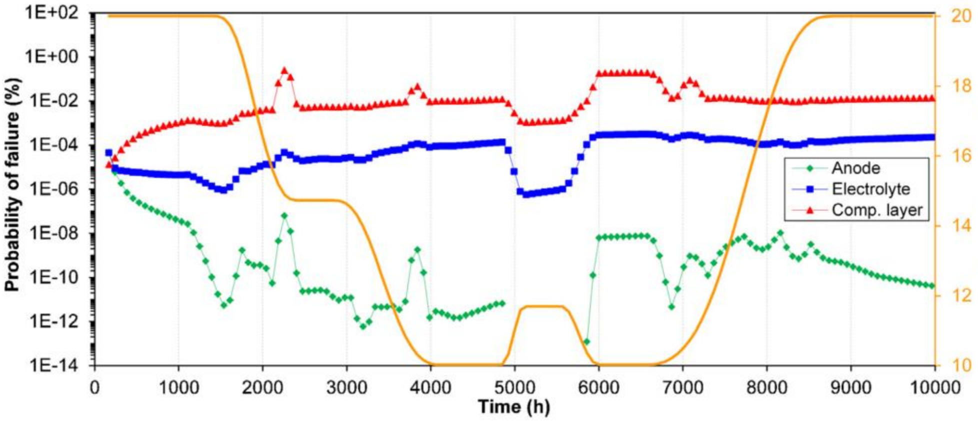

Figure 1 Probability of failure of the different layers of a SOFC stack as a function of time and changes in current intensity[1].

The conclusion of this that the creep rate and strength has a significant impact on the probability of failure, and must be characterized well. The purpose of WP2 is to determine the creep properties and strength of the main structural components of the SOFC, i.e. the anode support of the cell, the sealings, and the interconnector steel.

3.1.1 Mechanical properties of the cells

3.1.1.1 Strength

The strength of the TOFC anode supports were measured at every 200°C between room temperature (RT) and 800°C in the oxidized (NiO-YSZ) and reduced condition (Ni-YSZ). More than 30 samples from TOFC were measured at each temperature and condition.

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[1] F. Greco, H.L. Frandsen, A. Nakajo, M.F. Madsen, and J. Van herle, "Modelling the impact of creep on the probability of failure of a solid oxide fuel cell stack," *J. Eur. Ceram. Soc.*, **34** [11] 2695–2704 (2014).

The variation of elastic modulus (stiffness), Weibull strength and failure strain with temperature is shown in Figure 2. Based on this it was concluded:

- When in the oxidized all ceramic state, the stiffness and strength are insensitive to temperature variations.
- In the reduced state the Ni softens with temperature, lowering both the stiffness and the strength.
- The strength of the oxidized anode support is considerable stronger than the reduced. This is mainly attributed to the porosity, which increases with the reduction (NiO->Ni).
- The reduced anode support can be strained (elongated) more than the oxidized, and is thus more forgiving in terms of thermal gradients.

This data together with literature data formed the basis of the D2.1, and the achievement of M2.1.

Measurements of the strength of the anode at RT and 800°C of SOLIDpower's anodes were also completed (42 samples in total). The characteristic strength was lowered by approximately to the half at 800°C, compared to RT.

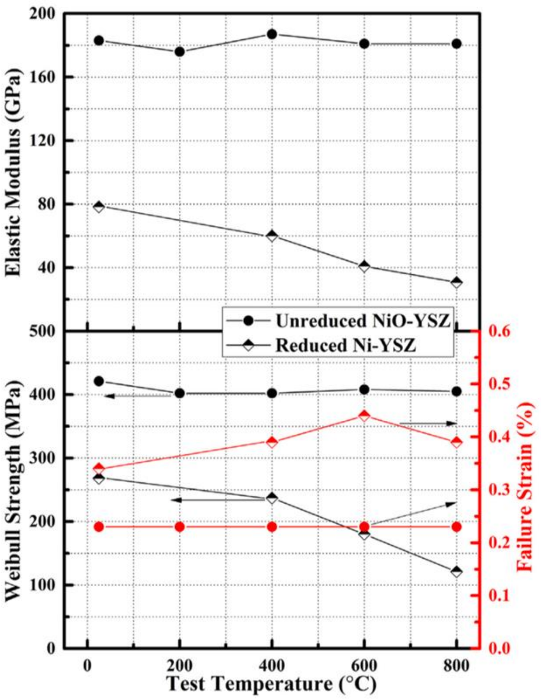

Figure 2 Weibull plots showing the distribution of strength for the reduced (a) and unreduced (b) Ni(O)-YSZ anode supports over the temperature range from room temperature to 800°C. All the Ni-YSZ specimens were reduced at 800°C in 6 h[2]

3.1.1.2 *Fracture Toughness of anode supports*

The fracture toughness of different anode substrate materials has been determined by double torsion test[3]. Results revealed the stress-activated phase transformation of 3YSZ can enhance the fracture toughness. NiO-8YSZ revealed a high-temperature thermo-mechanical stability and kept its fracture toughness even at 800 °C, however, with a still lower absolute value than that obtained for NiO-3YSZ (see Figure 3). Reduction of NiO-8YSZ leads to higher fracture toughness compared to the oxidized state due to the ductility of Ni.

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[2] D.-W. Ni, B. Charlas, K. Kwok, T.T. Molla, P.V. Hendriksen, and H.L. Frandsen, "Influence of temperature and atmosphere on the strength and elastic modulus of solid oxide fuel cell anode supports," *J. Power Sources*, **311** 1–12 (2016).

[3] G. Pećanac, J. Wei, J. Malzbender, Fracture toughness of SOFC anode substrates determined by a double-torsion technique, Journal of Power Sources, 327 (2016) 629-637.

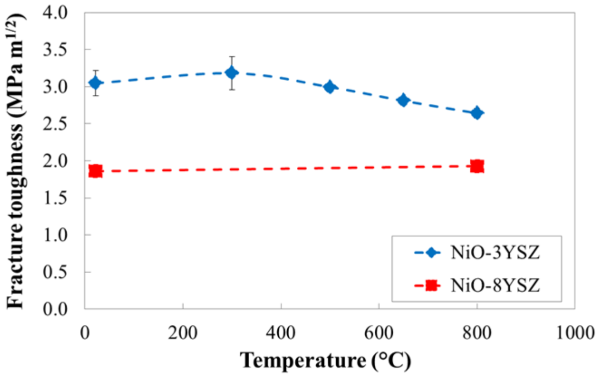

Figure 3 Facture toughness of Ni-3YSZ decreased at 800°C, while the values of NiO-8YSZ remained rather stable.

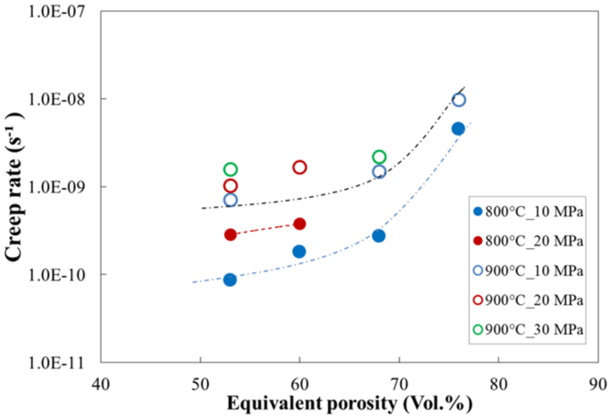

Figure 4 Creep rates as a function of equivalent porosities.

To measure the fracture toughness of anode support, single edge notched beam (SENB) method was also used. However, standard 3-point or 4-point bend loading of notched beams is not always suitable, especially for small or micro specimens (300 µm tall). Here a new approach is developed for calculating the fracture toughness from bending tests of notched beams[4]. The effect of notch geometry on the measurement of fracture toughness in any SENB test can be easily understood by using the concept of a visualized equilateral triangular zero-loading-bearing-volume (ZLBV) with its apex at the tip of the notch, or notch plus a defect extending from the tip of the notch.

To test such specimens load cells and displacement sensors of high sensitivity at low applied loads are needed. These are available in testing platforms such as instrumented nano-indenters. Here the elastic modulus and fracture toughness of thin cantilever beams of the anode support were measured using a micro-/nano-indenter. The Young's modulus and fracture toughness were determined to be 139 ± 4 GPa and 2.13 ± 0.27 MPa.m$^{0.5}$ respectively using this method[5].

3.1.1.3 Measurement of creep of the anode

Three common-used testing methods to assess the materials creep behaviour[6] (compression, ring-on-ring, four-point-bending). A diffusion-dominated creep mechanism of Ni-8YSZ was found for both compressive and tensile creep, while the activation energies show a dependence on the material´s composition, probably also related to temperature and loading modes. Porosity significantly reduces the creep resistance and yields larger creep rates (Figure 4).

Several models are available to predict the creep rates for porous materials. For the considered material composition, it appears that YSZ carries most of the load during creep. The same was concluded by a microstructural study in the project, where the creep response of the microstructure was modelled by a 3-D finite element model based on a reconstruction of the microstructure[7]. Comparing the numerical

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[4] X. Wang and A. Atkinson, "On the measurement of ceramic fracture toughness using single edge notched beams," *J. Eur. Ceram. Soc.*, **35** [13] 3713–3720 (2015).

[5] L.J. Vandeperre, X. Wang, and A. Atkinson, "Measurement of mechanical properties using slender cantilever beams," *J. Eur. Ceram. Soc.*, **36** [8] 2–6 (2016).

[6] J. Wei, J. Malzbender, Steady state creep of Ni-8YSZ substrates for application in solid oxide fuel and electrolysis cells, Journal of Power Sources, 360(2017) 1-10.

[7] K. Kwok, P.S. Jørgensen, and H.L. Frandsen, "Computation of effective steady-state creep of porous Ni-YSZ composites with reconstructed microstructures," *J. Am. Ceram. Soc.*, **98** [9] 1–8 (2015).

homogenization prediction and existing analytical models showed that the Ramakrishnan–Arunchalam creep model captures the relationship between creep factor and porosity most closely.

The primary and secondary creep of Ni-YSZ were also investigated using the developed metamodel parameter estimation method (see Section 3.1.6). Good agreement between measurements and a strain-hardening model predictions was found over the temperature range of 700-800°C. The measured temperature of the creep rate constant follows Arrhenius law with an activation energy in the range of 130 kJ/mol.

The measured thermo-elastic, creep and strength properties of the Ni(O)-YSZ were implemented in the WP4 stack thermo-mechanical model.

3.1.1.4 Finding of accelerated creep in anode supports

In the project a phenomenon, ‘accelerated creep’ in Ni(O)-YSZ anode supports, was discovered. The phenomenon takes place during the reduction of the fuel cell.

In Figure 5 the displacement over time at the center of an anode support sample in three-point bending is shown. The accelerated creep is resulting in the fast displacement at reduction (steep drop of curve). This occurs at the shift of atmosphere around the sample from air to a mixture of 9%H_2 in N_2, which reduces the sample.

The accelerated creep is approximately 10^4 times faster than creep during operation. The creep phenomenon is also seen to be concentrated in time, i.e. within 5-10 minutes. This corresponds to the estimated time for reducing the anode.

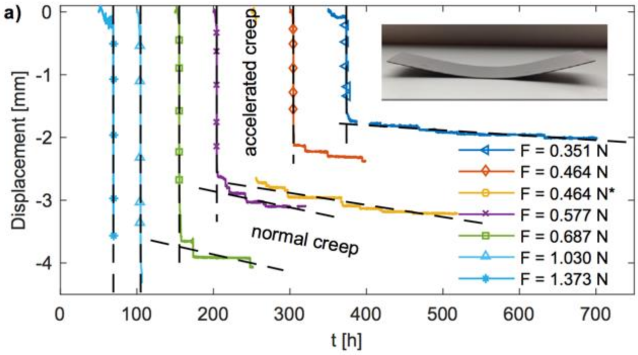

Figure 5 a) Displacement of the center of beams exposed to different loads in three-point bending and a shift of atmosphere from air to 9%H2 at 800°C (resulting in accelerated creep)[8].

Conclusively, it can be stated that the accelerated creep occurs at the combination reduction and loading, independent of loading method. As loading and reduction occurs at every SOFC stack assembly, this phenomenon has a significant impact stresses in the beginning of the SOFC stack life and onwards.

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[8] H.L. Frandsen, M. Makowska, C. Chatzichristodoulou, F. Greco, D.W. Ni, D.J. Curran, M. Strobl, L. Theil Kuhn, *et al.*, “Accelerated creep in solid oxide fuel cell anode supports during reduction,” *J. Power Sources*, **323** 78–89 (2016).

3.1.1.5 *Relaxation of stresses during SOFC stack assembly*

To investigate the impact of the accelerated creep phenomenon on a SOFC stacks, the stresses during heating, under reduction and during cooling was measured in-situ by use of X-ray diffraction (XRD). This was done both on at macro scale[9], i.e. studying the stresses in a half-cell (HC), and on a micro-structural scale[10].

The evolution of in-plane stress in the 8YSZ electrolyte was explored upon heating in air to different reduction temperatures of 600, 700 or 800 °C[9] (example at 700°C shown in Figure 6) Relaxation was found with some variations in results, but with one main conclusion: the stresses drastically decreases during reduction of the NiO to Ni at higher temperatures. This is attributed to fast chemically activated creep of Ni(O) at the typical reduction temperature.

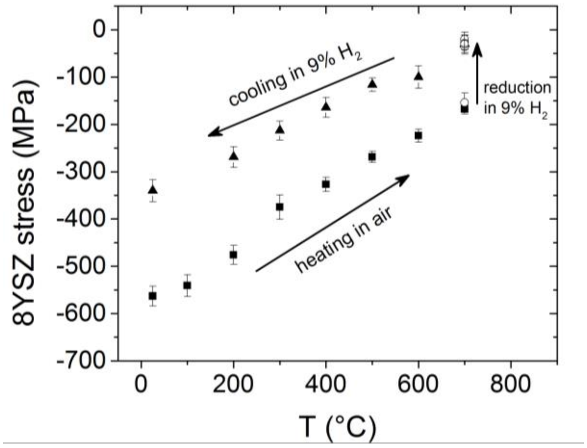

Figure 6 Evolution of in-plane residual stress in the 8YSZ electrolyte layer of the [9].

This entails that the stress in the SOFC practically is zeroed at the point of reduction. This has a great impact on the understanding of the stress evolution in a SOFC stack, both during assembly but also during reduction.

The anode material is being considered as a single layer, but is a composite of Ni and YSZ, with different physical properties. Thus, when cooling the composite from their sintering temperature, stresses will build up. YSZ will be in compression and the Ni in tension. In the project the micro-strain in each phase of the composite Ni(O)-8YSZ fuel electrode is determined by in-situ X-ray diffraction before, during and after reduction from the widening of the Bragg peaks due to local stress.

The Weibull strength of Ni-YSZ increased by 148 MPa (from 124 MPa to 272 MPa) when going down in temperature from 800 °C (after reducing at 800 °C), see Figure 2. This could also very well be due to increasing residual compressive stresses build up in the YSZ phase when going down in temperature, which was found in this micro-structural simulation to be -149 MPa[10].

3.1.2 Mechanical properties of the sealings

3.1.2.1 *Strength, elastic modulus and creep properties*

With respect to sealants, materials developed in Jülich, CSIC (Madrid, Spain) and SOLIDpower were characterized. The selected properties of elastic modulus, strength and creep were investigated.

H-Ag and H-F**[11]** sealant as a revised H matrix sealant with Ag particles and YSZ fiber reinforcements have been tested to observe the mechanical performance at room temperature and operating temperature[11].

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[9] H.L. Frandsen, C. Chatzichristodoulou, B. Charlas, R. Kiebach, K. Kwok, P. Norby, and P.V. Hendriksen, “In-situ determination of strain evolution in Solid Oxide Cells upon reduction. Part I: In-plane residual macro-strain in the cell layers,” (to be submitted).

[10] C. Chatzichristodoulou, K. Kwok, B. Charlas, R. Kiebach, P. Norby, P.V. Hendriksen, and H.L. Frandsen, “In-situ determination of strain evolution in Solid Oxide Cells upon reduction. Part II: Micro-strain within Ni(O) and YSZ in the fuel electrode,” (to be submitted).

[11] J. Wei, G. Pećanac, J. Malzbender, Mechanical behavior of silver reinforced glass–ceramic sealants for solid oxide fuel cells, Ceramics International, 41 (2015) 15122-15127.

Additional tests on 7.5 B(Ba) and 10 B(Sr) developed by CSIC[12] provide complementary results for the sealant study and comparison.

The SOLIDpower sealant revealed similar values of elastic modulus, hardness and fracture toughness as most of Jülich sealants as determined via indentation testing. The elastic modulus decreases with increasing temperature rather moderately up to 600°C, whereas above that temperature a stronger progressive decrease was visible.

Fracture stresses were tested by bending test for materials used in Jülich stacks. In particular, Ag particles as filler in the standard matrix material significantly enhance the fracture strength. A non-linear behaviour deformation was observed due to residual glassy phases. The results verified that, crystallization can enhance the fracture stress generally, while diffusion process of Ag appeared to contribute to a strength decrease by the introduction of micro-cracks.

Torsion strength tests of SOLIDpower sealant material indicates lower values compared to Jülich's sealants [13] at RT and 800°C (see Figure 7).

The SOLIDpower sealant appeared to have an apparent self-healing ability.

The creep behaviour of SOLIDpower sealant was investigated in temperature range of 530-630°C using compressive tests in air. The derived average activation energies of 220 ± 65 kJ/mol is ~2 times lower than the activation energy of as-sintered Jülich H-P sealant. The SOLIDpower sealant shows higher creep rates compared to anode substrate and typical Jülich sealants even at a lower temperature.

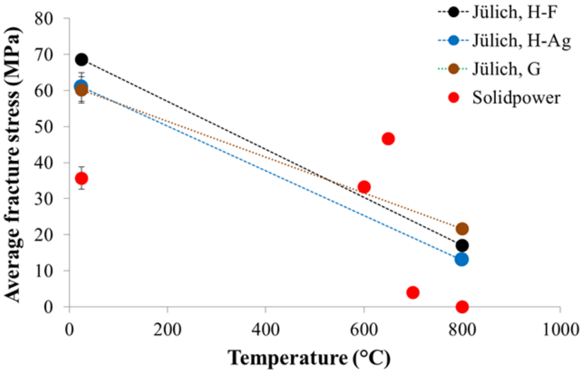

Figure 7 Comparison of the average shear stresses obtained from different sealants in as-sintered state at RT and 800°C.

3.1.2.2 Fracture toughness

The seal fracture energy was measured at room temperature using a wedge penetration test[14]. There is considerable scatter, and no clear trend of fracture energy with crack extension. The mean value of the fracture energy in the as-manufactured condition is 10.9 ± 1.8 J/m^2. The fracture faces indicate that the failure is mainly at the interface between the metal and its pre-formed oxide scale (formed during the first heat-treatment of the seal) along the interface between the seal and the upper (thinner) beam.

The fracture energy for the specimens after ageing for 1000 h is very similar to that of the as-received specimens, albeit with an increase in the mean value of the fracture energy, 15.3 J/m^2. The fracture surfaces show a transition with ageing from failure dominated by separation at the interface between the metal and pre-formed oxide, to separation within the oxide phases. There is also a change in the sealant in that it becomes coloured by interaction between the sealant and the metal (or pre-formed metal oxides).

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[12] S. Rodríguez-López, J. Wei, K.C. Laurenti, I. Mathias, V.M. Justo, F.C. Serbena, C. Baudín, J. Malzbender, M.J. Pascual, Mechanical properties of solid oxide fuel cell glass-ceramic sealants in the system BaO/SrO-MgO-B2O3-SiO2, Journal of the European Ceramic Society, 37 (2017) 3579-3594.

[13] T. Osipova, J. Wei, G. Pećanac, J. Malzbender, Room and elevated temperature shear strength of sealants for solid oxide fuel cells, Ceramics International, 42 (2016) 12932-12936.

[14] PROSOFC D2.2: Technical Report on Sealing Failure Mechanism,

3.1.3 Measurement of transient creep of interconnect steels

An experimental method for in-situ measurement of the transient creep response at operational conditions is developed[15]. The methodology utilizes a remotely installed length measuring setup involving laser micrometer is used to monitor deformations in the loaded samples in a furnace.

With his equipment primary creep strain, constant strain rate (Figure 8a), and relaxation (Figure 8b) can be recorded with high accuracy in-situ.

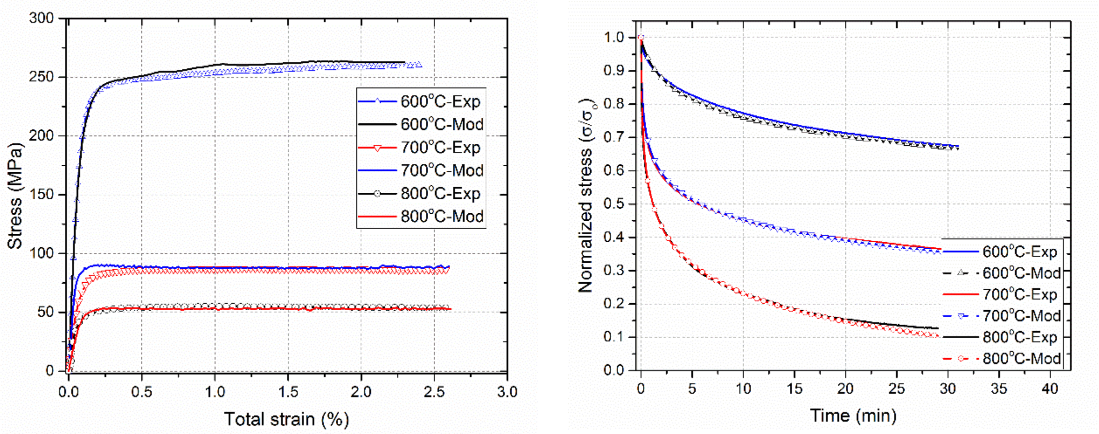

Figure 8 Comparison of experiments on Crofer 22 APU and model at different temperatures with (a) constant strain rate (b) relaxation behavior[16].

To utilize the measurements recorded by the high temperature creep measurement equipment in SOFC stack models, these must be described by a mathematical material model (constitutive law). It was thus investigated whether a further development of the so-called Chaboche's unified power law together with isotropic hardening can represent the transient behaviour of Crofer 22 APU[16]. A comparison between the model and the measurements are seen in Figure 8.

3.1.3.1 Abnormal creep behaviour of commercial interconnect steels

For SOFCs stacks different steel compositions are used by different manufacturers. The transient primary creep steel of the stack manufacturer in the PROSOFC project, SOLIDpower, was investigated and compared to the two commercial alloys Crofer 22 APU and Crofer 22 H from ThyssenKrupp.

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[15] T.T. Molla, F. Greco, K. Kwok, P. Zielke, and H.L. Frandsen, "Development of high temperature mechanical rig for characterizing the viscoplastic properties of alloys used in solid oxide cells," *J. Test. Eval.*, (2017).

[16] T.T. Molla, K. Kwok, and H.L. Frandsen, "Transient deformational properties of high temperature alloys used in solid oxide fuel cell stacks," *J. Power Sources*, **351** 8–16 (2017).

In Figure 9 the primary transient creep behaviour of the three different steels are compared with tensed with 12 MPa. Transient creep of Crofer 22 APU, is seen to creep significantly faster than the Crofer 22 H and the steel of SOLIDpower.

The steel of SOLIDpower also seems to have another unique feature; it contracts after half an hour to one hour of tensile loading. It could be due to release of residual stresses during the manufacturing or perhaps growth of laves phases during the first heating after manufacturing, but this deserves further research.

With some annealing (35 hours at 700°C), this effect can partially removed.

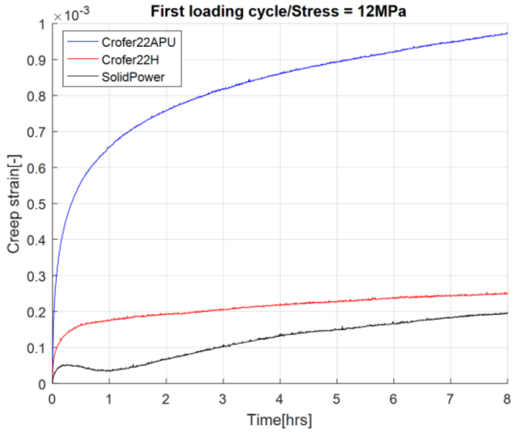

Figure 9 Transient primary creep at 700ºC of Crofer 22 APU, Crofer 22 H and the steel used by SOLIDpower.

3.1.4 Computational efficient modelling of creep of interconnects

Thermo-mechanical analysis of the entire solid oxide fuel cell (SOFC) stack at operational conditions is computationally challenging if the geometry of metallic interconnects is considered explicitly. This is particularly the case when creep deformations in the interconnect are considered in addition to elasticity. In the project this is addressed by using a mathematical abstraction of the actual geometry, i.e. homogenization. By this method the effect of the geometry is built into an “effective anisotropic material law” for a continuum block of material, which then represents the interconnect in the stack model.

This is done by a finite element model of a part of the actual repetitive geometry from this deduct a constitutive law for the homogenized structure (effective material law). Deformations involving the elastic, creep as well as effect of changes in the geometry due to contact is accounted for. The geometry was modelled by a 2D plane strain model is used to model. The mathematical material law (constitutive law) reminds of that of an anisotropic creep law, with a state variable accounting for the distortion of the microstructure.

In Figure 10, the developed constitutive law is verified by comparing its predictions for creep strain with results from the original 2D finite element model for different loading conditions. Further examples can be found in ref. 17. The constitutive law is found to describe the initial non-linear contacting as well as the linear steady state creep satisfactorily.

The result of this work is thus that a very complex geometry can be considered in a mechanical model of SOFC stack with tremendous computational gains and with a satisfactory description of the macro-scopic deformation in the stack.

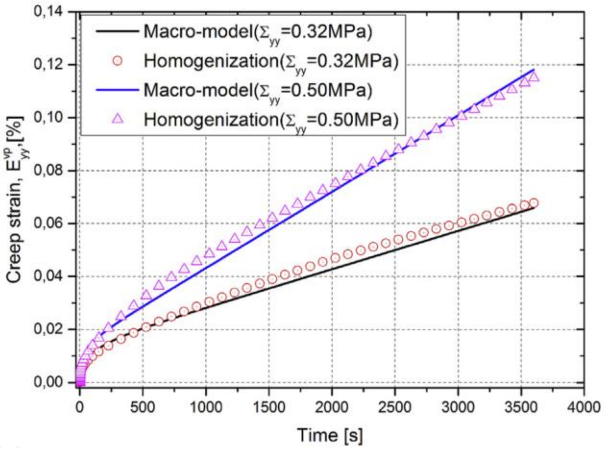

Figure 10 Comparison of average creep strains from the microstructural model and the homogenized model under load in the yy-direction[17].

3.1.5 Evolution of the thermo-elastic properties of the SOLIDpower electrode materials

Computational homogenization was also applied to investigate the effects of microstructural alterations on the thermo-elastic properties of the Ni-YSZ and LSCF-GDC electrodes (as with the interconnects in previous section). The studies were based on 3-D reconstructions obtained by FIB-SEM serial-sectioning of pristine and aged samples close to the YSZ electrolyte. The imaging methods applied in WP8 allow 3-D imaging at high spatial resolution (in the range of 10 nm; conditions were therefore set for isometric voxel ranging from 7-12 nm in the dataset), which enabled the detection of localized defects larger than this size (Figure 11).

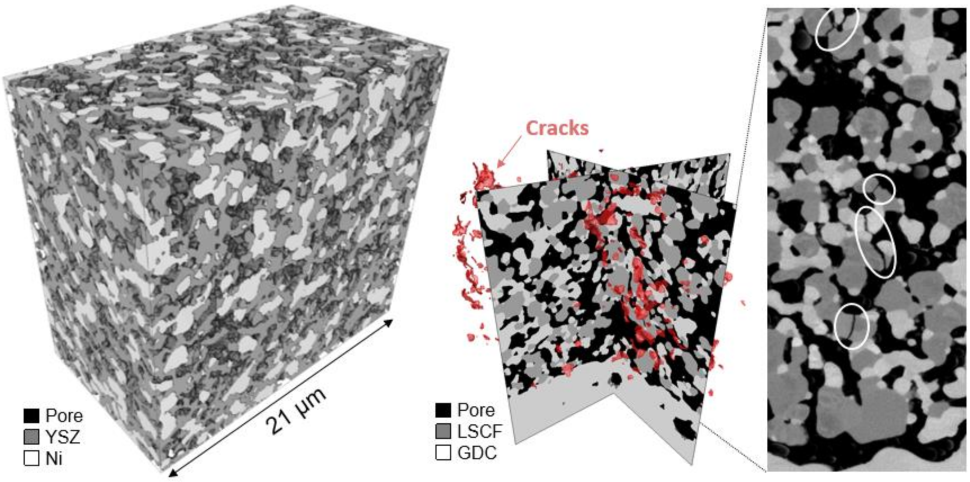

Figure 11 3-D view of the pristine Ni-YSZ (left) and LSCF-GDC (right) imaged by FIB-SEM serial sectioning, after segmentation. The close-up view shows cracks in a cross-section from the energy selective backscatter (EsB) detector.

In the Ni-YSZ electrode a clear differences in the Ni and pore phases, whereas significant alterations of the YSZ phase could not be ascertained after 4700 h of short-stack operation. While the transport and

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[17] T.T. Molla, F. Greco, K. Kwok, P. Zielke, and H.L. Frandsen, "Development of high temperature mechanical rig for characterizing the viscoplastic properties of alloys used in solid oxide cells," *J. Test. Eval.*, (2017).

electrochemical properties were noticeably affected, the effect on the elastic properties of the anode remained small and likely below the detection limits of macro-scale mechanical testing. The relationship between microstructural and effective thermo-elastic properties were analysed and the main effect was simply related to a slight change in porosity. The changes in the morphology and topology, quantified by the decrease of the coordination number and neck size distribution, contributed together to approximately one fourth of the decrease of the Young modulus caused by ageing.

Extensive microcracking was detected in the LSCF-GDC cathode (Figure 11). While a significant detrimental effect on the stack reliability is not expected, such damage can alter the electrochemical performance. The 3-D reconstructions were segmented including the cracks to examine relationships between micro-cracking and local morphology. Micro-cracks were intra and inter-phase. Most were found in the LSCF phase (41% of interfacial surface area), followed by GDC and LSCF/GDC interfaces. They caused a decrease in effective elastic modulus ranging from 2-15%, depending upon the analysed sample volume. An analysis based on surface curvature and neck and pore sizes suggested that weak locations can be identified a priori to some extent. The detected micro-cracks were indeed located on average close to regions of higher curvature and narrow necks.

3.1.6 Using the COPRD for improving measurement accuracy

To bridge the activities in WP2 and WP4 the COPRD was also used more accurately determine the strength and elastic properties of the SOLIDpower cells by 4-point bending.

Two setups for 4-point bending were built to characterize the strength and creep properties of the SOLIDpower cells, which allow testing at operating SOFC temperature and under controlled atmosphere. A metamodel-based parameter estimation method was used to obtain an improved accuracy by the quantification of the effects of intrinsic measurement errors. This also served as a development case for the COPRD optimisation workflow in WP4. Here a model was compared to the measurements, which allows accounting for material non-linearity and inaccuracies due to friction, anticlastic curvature and large deflection, among others during a complete loading-unloading cycle.

The analysis of the experimental results by metamodel parameter estimation revealed that friction between the sample and the rollers is problematic for ensuring accurate measurements of the elastic and creep properties, if standard data post-processing procedures are used. The elastic modulus of the Ni(O)-YSZ support was measured at room and high temperature in air (NiO-YSZ) or reducing atmosphere (Ni-YSZ, Figure 12). The decrease of the elastic modulus by approximately 22 % at 800°C (Ni-YSZ) is in line with computational homogenization calculations based on 3-D FIB-SEM serial sectioning reconstructions from WP8.

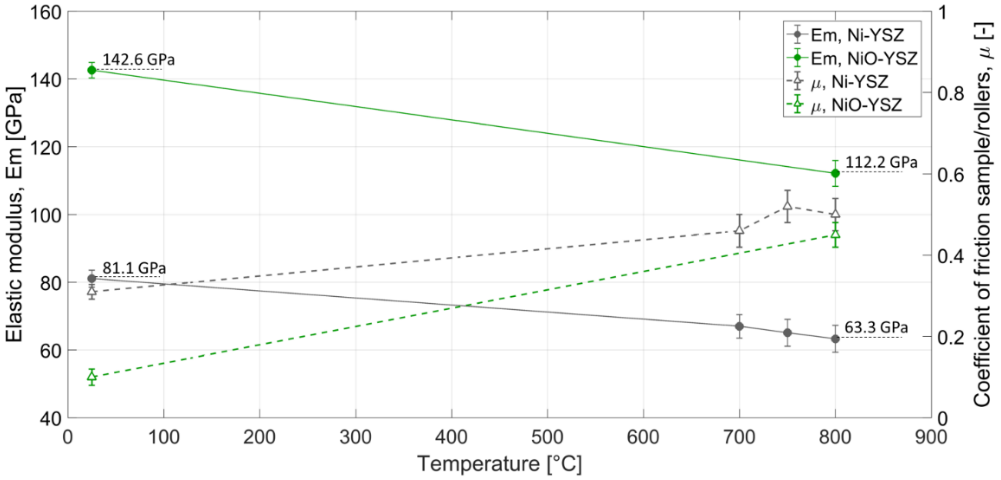

Figure 12 Estimated elastic modulus (filled circle symbols) and coefficient of friction between sample and rollers (empty triangle symbols). Estimated parameters correspondent to NiO-YSZ and Ni-YSZ samples are depicted respectively by green and grey colors.

3.2 Electrochemical testing, characterization and modelling

WP3 is related to the electrochemical testing, characterization and modeling, aiming at:

- detailed electrochemical analysis of baseline and selected redesigned cells
- evaluation of reforming kinetics of baseline and selected redesigned cells
- close to reality tests for segmented single cells with determined temperature distribution
- microstructure analysis and reconstruction of baseline and selected redesigned cells
- complex continuum models of electrochemistry and reforming catalysis for stack and system simulations and probabilistic design optimizations,
- validation of the improvements by statistically ensured results of performance, reproducibility and durability tests

The work in WP3 is related to 6 tasks, the results are summarized below.

Task 3.1: Statistically ensured performance testing of single cells

This task was solely carried out by TOFC. At least one cell of every cell batch produced at the TOFC pilot production facility is tested in a quality inspection test unit which operates in a 24 h cycle. The tested cells are picked randomly from the production lines before the final sintering of the cathode contact layer which thus is green at the start of the test. In total 44 cells have been tested over a period of roughly 6 months covering fall 2013 to spring 2014. In addition, cells from a reference cell batch are measured at regular intervals to ensure stable operation of the QA cell test. The measurements showed batch to batch variation in the ASR below 20 mΩ·cm^2. The results could be confirmed in stack tests.

Task 3.2: Detailed electrochemical analysis

TOFC and SP cells were tested by means of electrochemical impedance spectroscopy according to validated procedures. Operating conditions (temperature, gas composition at cathode and anode, current density) were varied to determine the parameter dependencies of the individual electrochemical processes in the cell. Whereas this approach worked fine for the TOFC-cells, the testing of ASCs of the new partner SOLIDpower caused significant problems, which had not been observed with other ASCs before. The problems were related to a bending and deformation of the cell during testing, which resulted in improper contacting and severe leakages. The results showed that the SP-ASC exhibits electrochemical properties and loss mechanisms that are qualitatively similar to other anode supported cells. Measure to improve the cells stability by modifications in the test bench and sealing were not successful, a reliable testing of the SP ASC was not possible.

Task 3.3: Analysis of the reforming kinetics

A number of TOFC ASCs were successfully tested with different fuel gases. Durability tests with durations exceeding 1000 h were performed in the internal reforming mode and fuel utilizations up to 80%. Even under these harsh conditions the cells showed a high performance (average power density > 400 mW/cm²) and stability.

Task 3.4: Close to reality tests

The purpose of the close-to-reality tests is to simulate temperature fields for single cell tests as they occur in a real stack. The results have been used for calibration of the stack model's chemistry, electrochemistry, and heat management calculations.

A number of tests in H_2 operation with high temperature gradients were performed to calibrate the thermal behavior of the AVL CFD simulation model in WP4. The simulation model was enabled to simulate the temperature distribution for the complete segmented cell test equipment with only a few K difference between the measured and simulated temperature values.

Furthermore, a comprehensive test series was completed with tests in H_2 and CH_4 operation at 700, 750 and 800 °C to provide information about the temperature distribution, gas composition and local cell voltages for each segment. The data was used for the CV curve calibration and for the calibration of the reforming kinetics and for the CFD model in WP4. It could be shown that this experimental support for calibrating the CFD model is a very powerful approach as it provides more detailed and precise data on temperature distribution, gas conversion and cell voltages along the cell under realistic conditions compared to what can be measured in real stack tests.

please add a picture

Task 3.5: Microstructural analysis and reconstruction

To support the partner FZJ in the development of a mechanical creep model for anode supported SOFCs reconstructions of an anode supported cell have been provided. The reconstruction process was further developed to obtain an accurate reconstruction of the anode microstructure and ensure a reliable separation of nickel and YSZ. The improved methods were used to reconstruct larger parts of the anode functional layer and the anode substrate in order to ensure representativeness of the reconstructed volume.

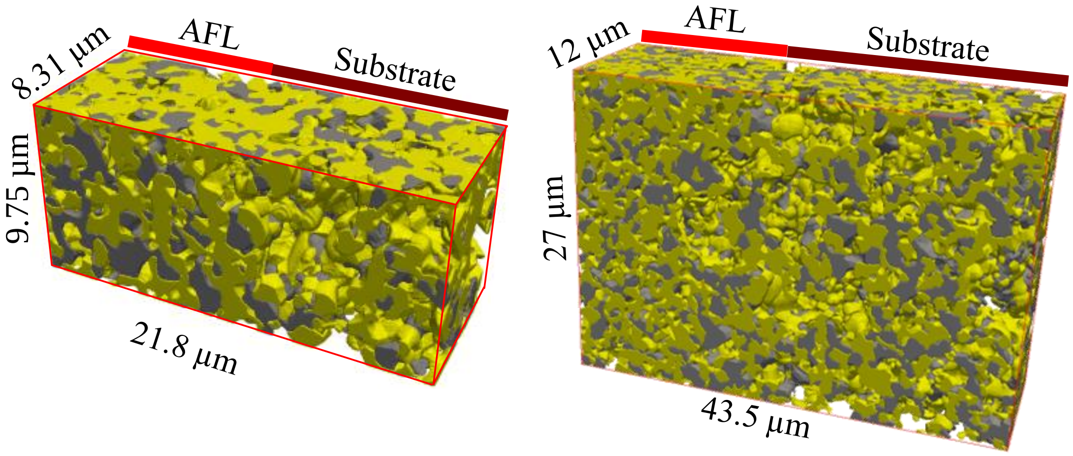

Figure 13: Reconstructed volumes of two Ni/YSZ anodes from FZJ containing the AFL and an adjacent part of the substrate.

Task 3.6: Electrochemical modeling

To set up an applicable equivalent circuit model and to evaluate starting values for an CNLS-fit the distribution of relaxation times for the various parameter variations were analyzed. As the cell is composed of similar materials and components as the anode supported cell developed at the research center Jülich, the existing model[18] could be adapted to describe the electrochemical properties of the TOFC cell.

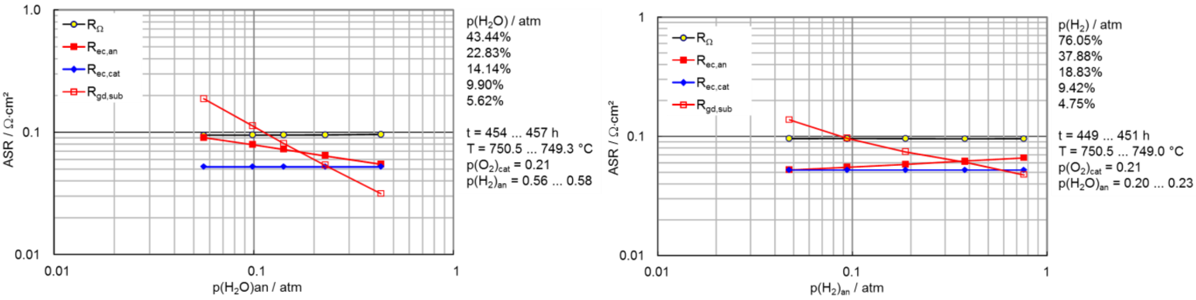

Figure 14: TOFC-cell, ASR-values of the different loss mechanisms – variation of pH2O,anode and pH2,anode

The investigated TOFC cells show qualitatively similar dependencies of the individual loss mechanisms on the different operating parameters. From a quantitative point of view there are of course differences related to differences in applied raw materials, cell production methodologies and resulting electrode microstructures. In the first part of the project (M1 to M18) a model for the TOFC-ASC could be set up. A similar approach was intended to be applied for the SP-ASC. As the cells showed severe problems with respect to their flatness, resulting in an insufficient contacting, a partial reoxidation and a subsequent cracking of the substrates, reliable measurements were impossible and the required model parameters could not be evaluated.

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[18] A. Leonide, V. Sonn, A. Weber and E. Ivers-Tiffée, "Evaluation and modeling of the cell resistance in anode-supported solid oxide fuel cells", J. Electrochem. Soc. **155**, p. B36-B41 (2008).

3.3 Assessment and modelling of baseline design under consideration of stochastic properties.

WP4 is related assessment and modelling of baseline design under consideration of stochastic properties. Its aims are to:

- Provide a methodology for 3D description of spatial distribution of material properties based on a random field model
- Relate the probabilistic model to the experimentally obtained properties of base materials
- Establish a statistical relationship between the material properties and the most relevant load effects in view of potential damage/failure based on multi-physics simulation
- Reduce the number of random variables required for a sufficiently accurate description of the component and system reliability
- Provide both a physical model based as well as a purely statistically based description of the production process including the effect of non-destructive testing and screening
- Establish computationally efficient multi-physics meta-models of stack conditioning and operation to be utilized for automatic design variation and cost-based design optimization

The work in WP4 is related to 7 tasks, the results are summarized below.

Task 4.1 Methodology of 3D description of the spatial distribution of material properties and loading conditions in terms of random fields

In order to address the specific needs of the thermo-chemical analysis, special focus has been placed on the tools to analyse one-dimensional random fields (or ”signals”). To this end, the software packages optiSLang and SoS have been enhanced with functions to deal with one-dimensional random fields specifically. Moreover, the visualization tools available in SoS have been further enhanced for a clear representation of signal, either defined pointwise or along a continuous (space or time) axis.

Task 4.2 Quantitative calibration of random field models based on available statistical data from tests

The question of identifying material parameters from experimental results has been investigated (WP2). For this purpose, a four-point-bending test was simulated, and the quality of the input-output relations (signifying the accuracy of the mechanical model) was tested using optiSLang. For this purpose, a 3-D finite-element continuum model was implemented in ABAQUS.

Non-linear material properties can be implemented in the model. The simulation of the 4-point bending experiment for the measurement of the Young’s modulus requires only linear elasticity, whereas material non-linearity is needed for the estimation of the primary and secondary creep parameters. Since the relative motion between the sample and the rollers is assumed potentially non-negligible, the large-sliding contact tracking is adopted in the model.

Task 4.3 Multi-physics simulation at stack level considering stochasticity

AVL established a full-scale stack simulation based on the real geometry of the SOLIDpower stack. The local temperature phenomena (e.g. temperature distribution of cells in the stack center vs. cells on top and bottom of stack) were investigated in more detail. Eventually the stack simulation was operated under H2 and CH4 conditions. Especially, the implementation of the reforming kinetics into the full-scale stack model was an important step in this task. The AVL FIRE CFD simulation model of the full stack was setup based on the geometry data provided by SOLIDpower.

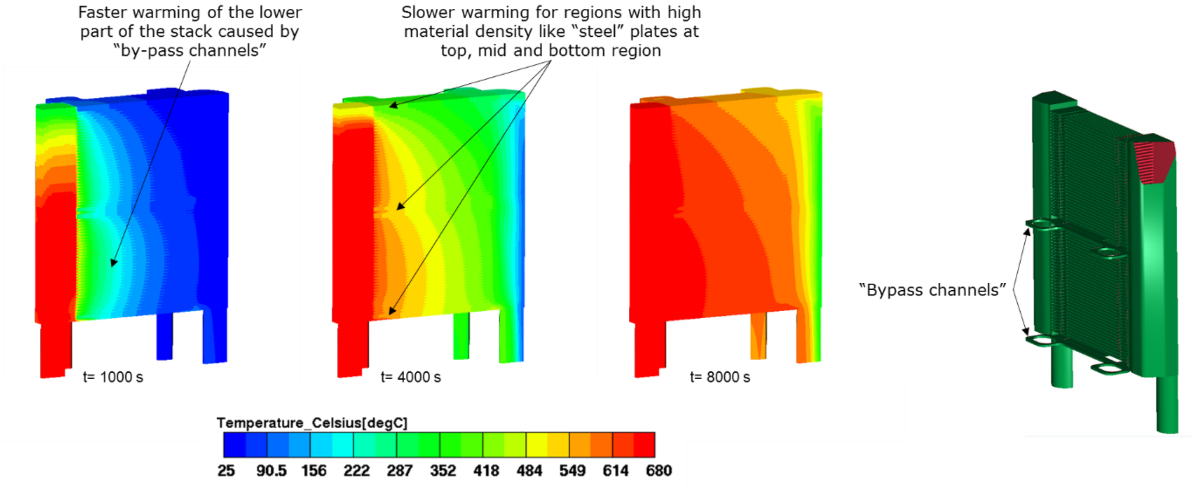
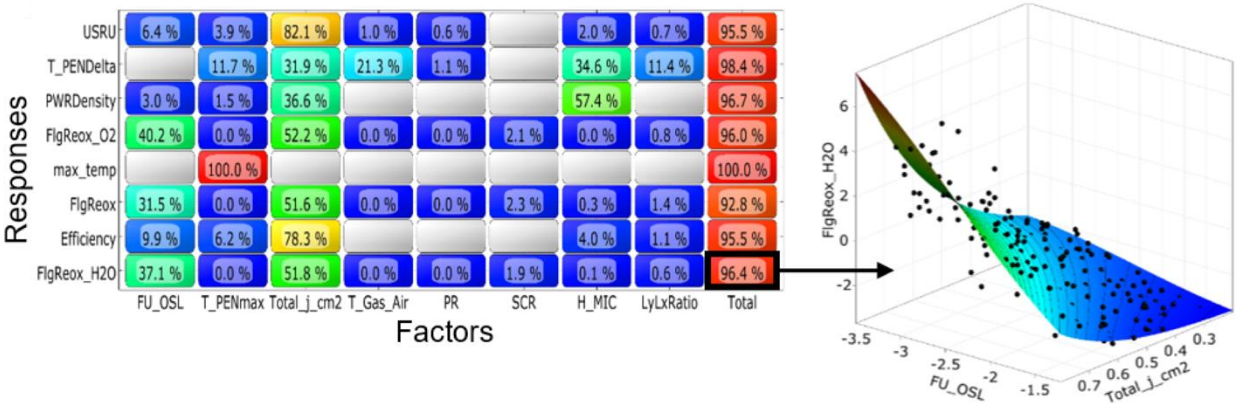


Figure 15: Stack model, temperature distribution warm-up

Out of the full stack model with 66 cell layers first a smaller CFD model (3 cell model) was created in order to calibrate parameters for the SOFC module of AVL FIRE CFD code in a faster way. Detailed results on this task are reported in D4.3. The full-scale and 3-cell model were the basis for the optimization task 4.6 by using optiSlang.

Task 4.4 Model order reduction regarding random field representation based on stochastic sensitivity

Sensitivity analysis is most efficiently carried out using simplified representations of the physical processes in terms of so-called metamodels. The quality of a metamodel is evaluated by the coefficients of prognosis (CoP). Typically, a CoP equal or better than 90 % is considered as the threshold for robust optimization. The continuous flags for the reoxidation of nickel reached a CoP of 96%, instead of 59% in initial attempts (Figure 6). The change of the variable for the fuel utilization allowed to increase the number of successful samples at high fuel utilization, so that its influence on the efficiency could be better captured. Moreover and as expected, the fuel utilization had after re-scaling an effect on the power density and voltage, and a larger effect on the risk of Ni reoxidation flags.



	FU_OSL	T_PENmax	Total_j_cm2	T_Gas_Air	PR	SCR	H_MIC	LyLxRatio	Total
USRU	6.4 %	3.9 %	82.1 %	1.0 %	0.6 %		2.0 %	0.7 %	95.5 %
T_PENDelta		11.7 %	31.9 %	21.3 %	1.1 %		34.6 %	11.4 %	98.4 %
PWRDensity	3.0 %	1.5 %	36.6 %				57.4 %		96.7 %
FlgReox_O2	40.2 %	0.0 %	52.2 %	0.0 %	0.0 %	2.1 %	0.0 %	0.8 %	96.0 %
max_temp		100.0 %							100.0 %
FlgReox	31.5 %	0.0 %	51.6 %	0.0 %	0.0 %	2.3 %	0.3 %	1.4 %	92.8 %
Efficiency	9.9 %	6.2 %	78.3 %				4.0 %	1.1 %	95.5 %
FlgReox_H2O	37.1 %	0.0 %	51.8 %	0.0 %	0.0 %	1.9 %	0.1 %	0.6 %	96.4 %

Figure 16: Matrix of the coefficient of prognosis; showing in particular the improvement of the metamodel for the Ni reoxidation flags.

Task 4.5: Development of stochastic models for process parameters in manufacturing

A database containing stack production monitoring data was set up in WP6. It collects production parameters from SOLIDpower standard production, as well as resulting indicators for the quality of the product such as electrochemical performance, gas tightness, etc.

It was then evaluated if stochastic models could be established for selected quality indicators, using the tools offered by Dynardo. A particular complication arose from the fact that production parameters in this particular real production environment at SOLIDpower are only seldom varied as continuous variables, but rather undergo discrete changes such as changes of recipes, modifications of designs, change of materials or batches of components.

It was found in particular that those discrete changes impacted the manufacturing quality in far larger extent than continuous changes performed in that particular context and period of development of the product.

Task 4.6 COPRD design optimization

A workflow using the optiSLang DOE software tool was used to optimize the warm-up procedure of a Solid Oxide Fuel Cell Stack by analyzing results of 3D CFD simulations (AVL FIRE). For a defined 3D CFD half-model (symmetry) of a full SOFC stack, basic warm up conditions were defined. These definitions served as basic parameters which were finally optimized. The optimization procedure was performed with the meta-model in order to determine the optimal warm up parameters. As a last step the optimized parameters are reevaluated by a final CFD simulation. This work flow was used for meta modeling done by optiSlang.

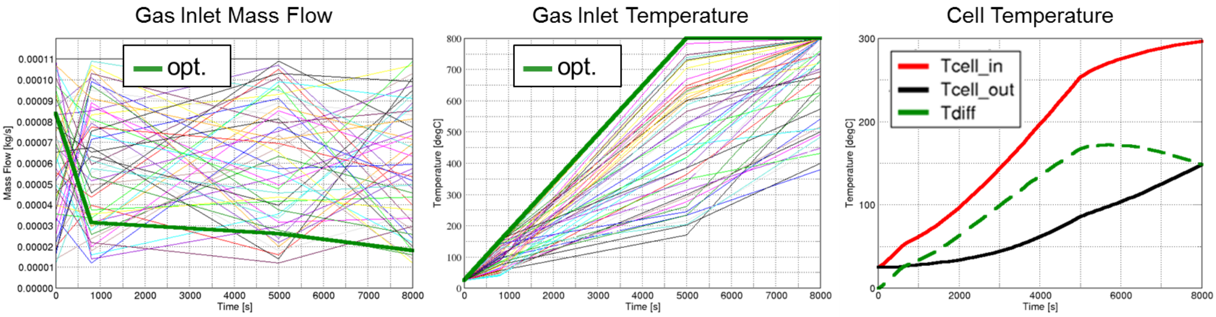


Figure 17: Optimization results (mass flow, temperatures)

The overall results look quite reasonable. A very basic workflow for optimizing SOFC stack warm up using DOE software optiSLang, could be established.

Task 4.7: Comparison of first and second stack generation

A full scale stack CFD simulation model, based on the real geometry of the SOLIDpower stack, was setup in AVL FIRE. Due to symmetry, only the half of the stack model was used for the simulations. During the simulation approach the full scale stack model was improved regarding the material parameters and compared with the 3 cell model. The calibration of the full scale stack simulation was based on real operating parameters (H2 operation) from SOLIDpower. Therefore, the CV curve of the simulation can be compared with the real measured CV curve from the test. The results show good correlation with the real test.

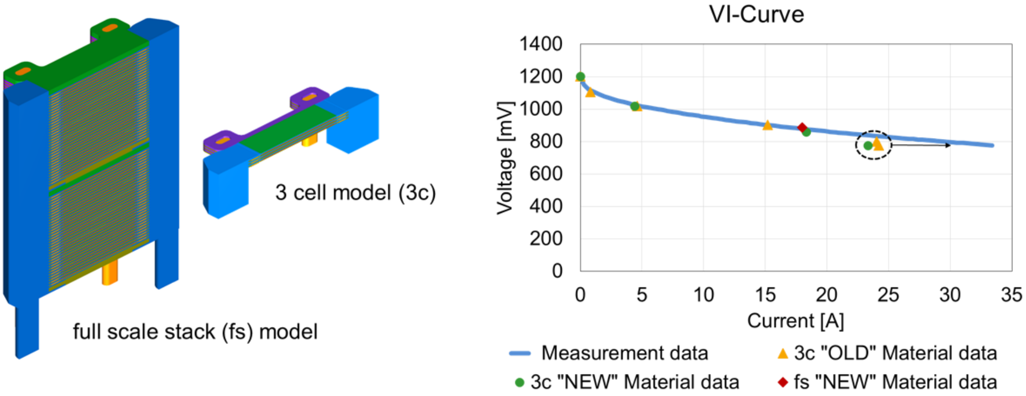


Figure 18: Results of the full scale stack (fs) model and the 3 cell model (3c) with old and new material parameters

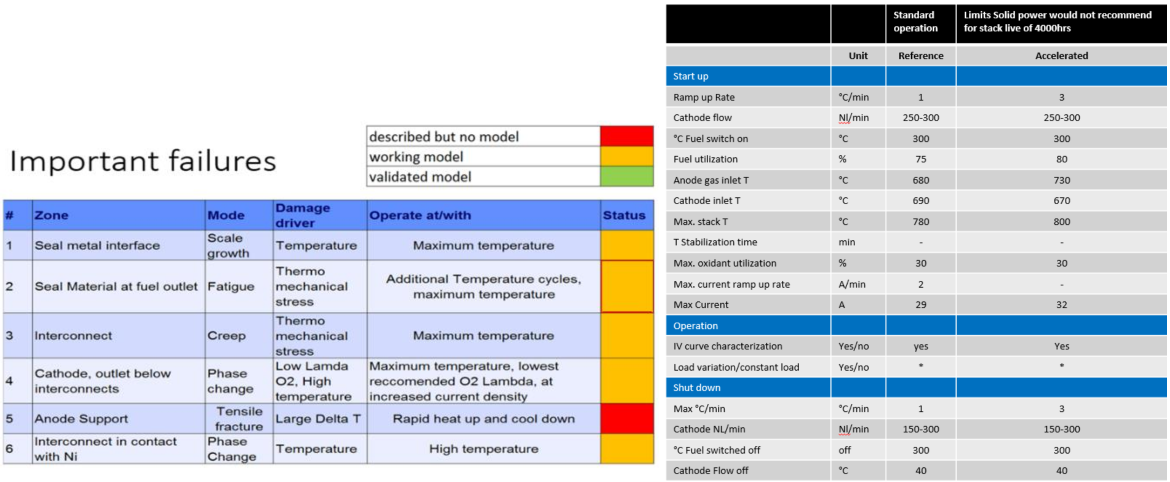
3.4 Reliability targets and testing procedures

The objectives of the work package WP5 are given here in a list.

- Definition of base case design and reference application
- Identification of main failure modes and mechanisms
- Definition of reference load cycle and reliability targets
- Elaboration of the reliability test program

Task 5.4 Reliability test program (AVL)

The test program was defined in the midterm report in 2014. Since SOLIDpower entered the project and agreed to the suitability of the test program. Most failure modes were modelled and reported already in the 1st midterm report. The failure modes deemed valid for a validation test program have been updated in a second development loop in order to try to encapsulate the failure mode physics in a more detailed fashion, see Figure 19. Four failure modes were deemed as more relevant for the SOLIDpower stack in comparison to the TOFC stack and therefore developments were focused on these themes.



Important failures

described but no model	🟥
working model	🟧
validated model	🟩

#	Zone	Mode	Damage driver	Operate at/with	Status
1	Seal metal interface	Scale growth	Temperature	Maximum temperature	🟧
2	Seal Material at fuel outlet	Fatigue	Thermo mechanical stress	Additional Temperature cycles, maximum temperature	🟧
3	Interconnect	Creep	Thermo mechanical stress	Maximum temperature	🟧
4	Cathode, outlet below interconnects	Phase change	Low Lamda O2, High temperature	Maximum temperature, lowest reccomended O2 Lambda, at increased current density	🟧
5	Anode Support	Tensile fracture	Large Delta T	Rapid heat up and cool down	🟥
6	Interconnect in contact with Ni	Phase Change	Temperature	High temperature	🟧

		Standard operation	Limits Solid power would not recommend for stack live of 4000hrs
	Unit	Reference	Accelerated
Start up			
Ramp up Rate	°C/min	1	3
Cathode flow	Nl/min	250-300	250-300
°C Fuel switch on	°C	300	300
Fuel utilization	%	75	80
Anode gas inlet T	°C	680	730
Cathode inlet T	°C	690	670
Max. stack T	°C	780	800
T Stabilization time	min	-	-
Max. oxidant utilization	%	30	30
Max. current ramp up rate	A/min	2	-
Max Current	A	29	32
Operation			
IV curve characterization	Yes/no	yes	Yes
Load variation/constant load	Yes/no	*	*
Shut down			
Max °C/min	°C/min	1	3
Cathode NL/min	Nl/min	150-300	150-300
°C Fuel switched off	off	300	300
Cathode Flow off	°C	40	40

Figure 19: List of failure modes and damage drives relevant for the SOLIDpower stack & Reference and accelerated cycle

To make specific statements about the demonstrable reliability for a durability target of thirty thousand hours, a high acceleration factor and a long measurement duration are essential. In Figure 19 this dependence is shown. For a short measurement duration of three times 4000 hours a very high acceleration factor (≥20) is needed to make meaningful statements about the demonstrable reliability. For a longer test duration of three times 8000 and 16000 hours an acceleration factor of 10 and 5 is required. For the considered damage models in the PROSOFC project, the acceleration factors are located between one and three and the measurement on the test stands were 4000 hours for three different stacks. Therefore it is no possible to make precise statements about the reliability of the stack.

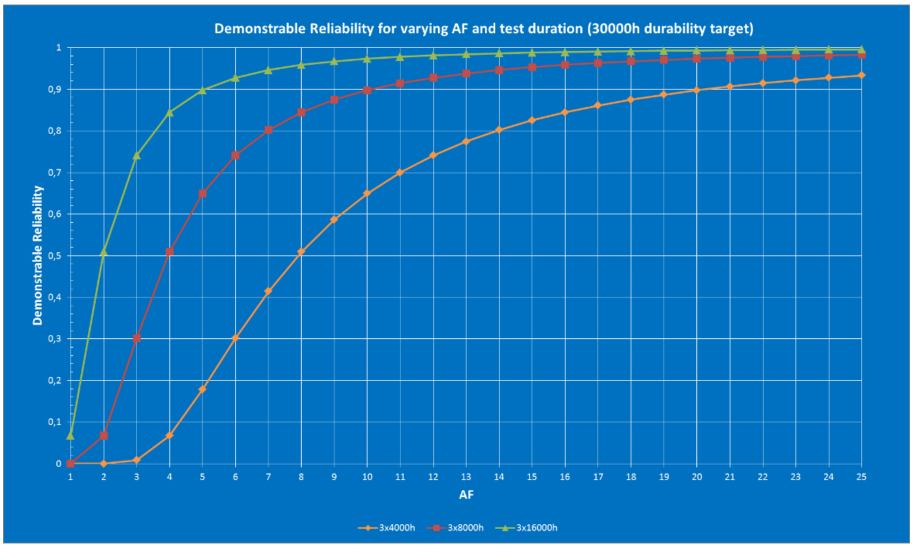


Figure 20: Demonstrable reliability in dependence of the AF for durability target of 30000h

In Figure 21 the demonstrable reliability is shown in dependence of the reliability target for an acceleration factor of three. For an acceleration factor of 3 which was a typical result from the damage model calculations, a measurement duration of three times forty thousand hours is needed to reach the B5 target (demonstrable reliability 0.95 for reliability target of 30000 hours).

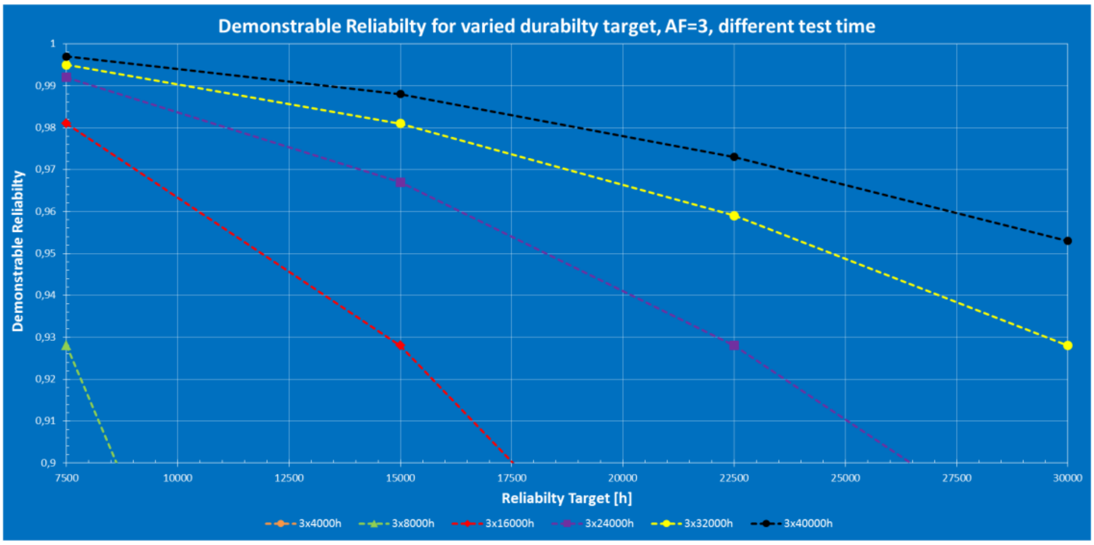


Figure 21: Demonstrabel reliability in dependence of the reliability target for an AF of three

The demonstrable reliability for the real test duration, three times four thousand hours, is so low that it cannot be shown in the graphic below. Even the highest calculated AF (AF≈3) from the examined damage models is much too low to make accurate predictions about the reliability of the stack (B5 target) for a measurement duration of three times four thousand hours. Another theoretical calculation was performed for a higher estimated acceleration factor of ten, Figure 22. Even with an increased AF for a damage model the measurement duration of three times four thousand hours is still too low to give precise information about the demonstrable reliability. It was calculated, that for an AF of ten for one of the damage models a measurement duration of three times 11605 hours would be needed to reach the B5 target.

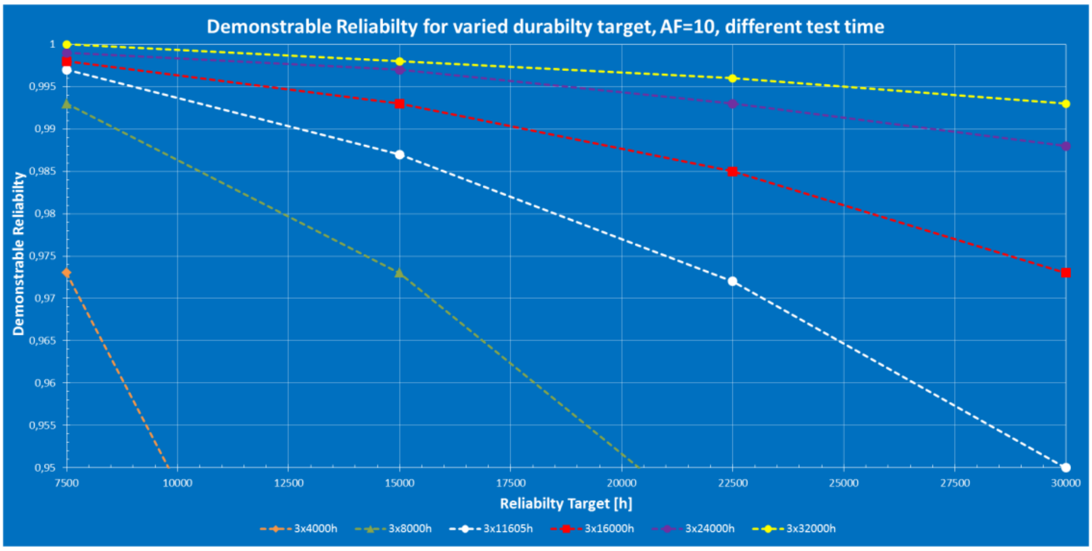


Figure 22: Demonstrabel reliability in dependence of the reliability target for an AF of ten

These values are far away from the achievements of the PROSOFC project with an AF of 3 and a measurement duration of three times four thousand hours.

Conclusions and recommendations

- 3 X 4000 hours is not enough to meet B5 Target if predicted AF are correct
- No determination possible for AF validation, because no post mortem analysis on stacks performed
- More realistic would be 3 X 11.6k hours testing for AF of 10 to reach B5 target
- Basis for future validation test program design and modification established
- Validation test recommendation completed

The most relevant failure models could be identified mutually in the consortium. Most failure models are developed and a stack test program was established. The final validation of the failure modes could not yet be performed as the stack test results are not yet available. From the stack test in WP7 the real operating parameters over time for the damage model calculation will be provided to finally estimate the acceleration factor achieved by the planned 4000 hr test cycles. The analysis will further identify the scale of the critical failure modes. This information can be used to consider and model the most critical failure modes in a FE simulation in WP4 which is based on the simulated temperature fields.

3.5 Improvement of cell design and manufacturability

WP6 is addressing the improvement of the stack parts and design for improved robustness, manufacturability and cost. The original plan is addressing the technology of TOPSOE stacks, focusing principally on the cell technoglogy:

- Substantially improve robustness, manufacturability, efficiency and cost of production cells.
- Establish enhanced production QC methods
- Increase the learning from available production QC data
- Provide cells with varied production history for measurements in WP2 and WP3
- Conduct production campaigns to establish progress and provide cells for stacks in WP7
- Provide production data for documentation of the project outcome

As the project was transferred to SOLIDpower/HTCeramix, the focus was set on the stack mechanical design in general, and on seal materials and construction in particular.

Task 6.1 Establishment of baseline manufacturing performance data structure and share baseline information data at project start (HTc, DTU)

To assess the baseline manufacturing performance, the stack production data has been introduced in a database going back to 2014. This dataset contains the key parameters for the stack sintering (flows, temperatures, etc.), as well as selected key performance indicators (KPIs) covering both stack performance (electrical power, efficiency, nominal voltages) and reliability indicators (hot and cold leak rates, quality of contact, homogeneity, etc.). The database contains further quality data about single stack components such as the cells.

The database is completed by a web-based interface that allows users to acces the detailed data of each produced stack, as well as to get statistical data and its evolution with time. Different indicators are available for analysis, such as the gas tightness, performance during the qualification run, etc. The tool is now permanently used for production monitoring.

This database has allowed to show statistically that the stack design improvements developed in the project have resulted in a major improvement of the quality and reliability of seals, resulting in improved gas tightness and thermal cycling capability.

T6.2 Design and material modifications based on SOLIDpower current technology and feedback from WP2, 3 and 4. (HTc, EPFL, DTU)

A synthesis was made of the results obtained in WP2, 3, 4 and 8, combining both experimental results on materials and interfaces, as well as simulation outputs and post-experiment analyses to understand the main stack failure modes and their origins. This synthesis lead to several design and material adaptations. In total, 9 stack design variants have been prototyped, among them 3 main variants during the last reporting period (referred to as A, B and C).

The thermomechanical simulations performed at EPFL (WP4) have explained to a large extent the possible seal failures observed on the original stack design. During the sintering and the first phase of operation, an important relaxation of stresses occurs in the glass-ceramic seals, both on the cell edges and in the fuel manifolds. If the stack is thermal-cycled, a situation arises where the glass seals are exposed to shear and, worse, tensile stresses, that can lead to delamination or failure. This conclusion enabled to redesign the seal system of the stack in-depth.

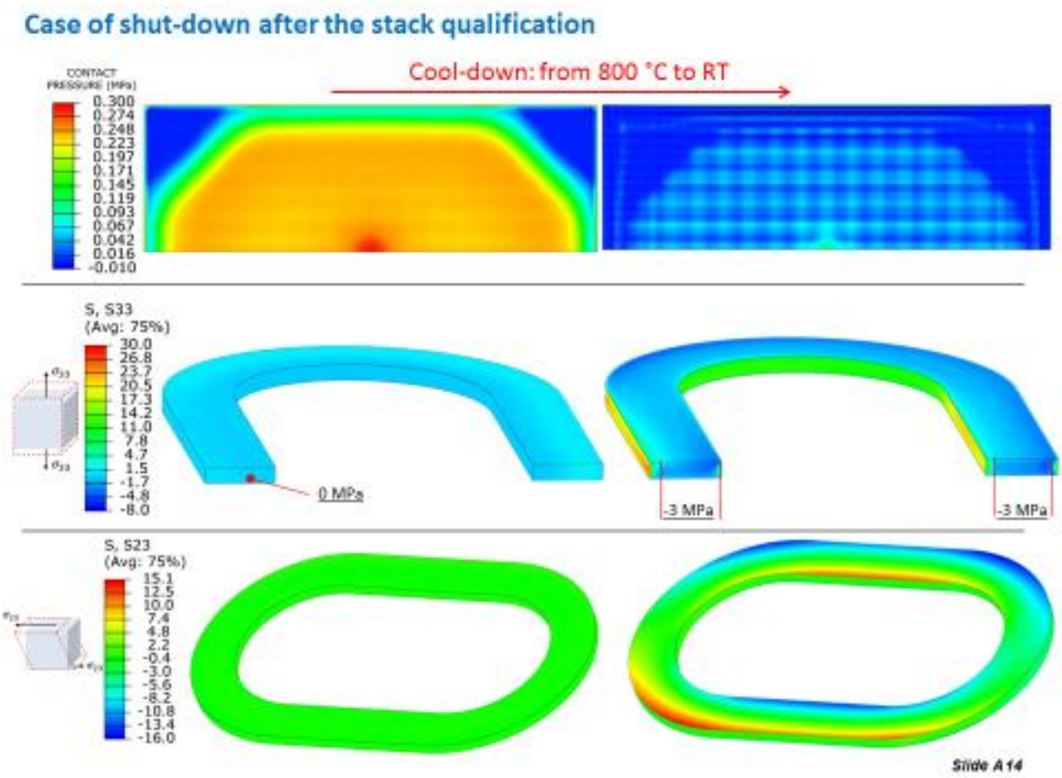


Figure 23: Evolution of thermomechanical stresses upon cooldown.

Material characterizations, in particular creep tests made at DTU (alloy) and FZJ (seals) showed that the selected materials were corresponding to the needs and that no further changes were immediately required from a thermomechanics point of view.

In the first two principal design modifications (A and B), the cell edge seal design, the fuel manifold seal designs and the stack end-plate designs were modified based on these findings. The last design iteration C was further addressing cost reduction for mass manufacturing. This design is currently been transferred in production for pre-series production and validation.

Task 6.3 Production campaign and testing on second generation of stacks integrating design changes (HTc)

The designs A, B and C have been put in production in 2017 and resulted in immediate improvements in some quality indicators, while keeping electrochemical performances at the same level. In particular, the gas tightness improved by one order of magnitude, as shown in the figure below.

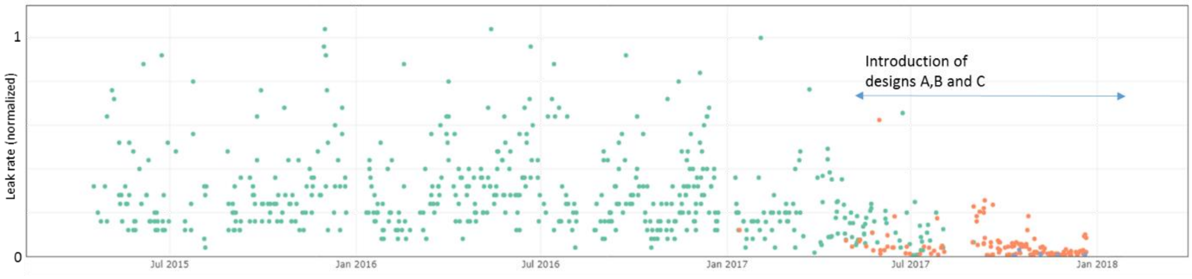


Figure 24: Improvement in gas tightness during the last years (normalized)

Not only this aspect was improved, but the thermal cycling capability was impacted as well. One stack of design B was successfully tested over 100 deep thermal cycles and was finally stopped for post-operation analyses. The thermal cycles were made in steam-reforming conditions simulating typical system operation context. With earlier designs, this number of thermal cycles was never reached on a full-scale stack.

The open circuit voltage of the stack experienced limited decay over the number of cycles, starting to degrade visibly only after about 90 cycles. The stack voltage at full load decreased in average by -29mV per cycle, which represents a decay of about 0.05% per cycle (voltage based). The result is shown in Figure 25.

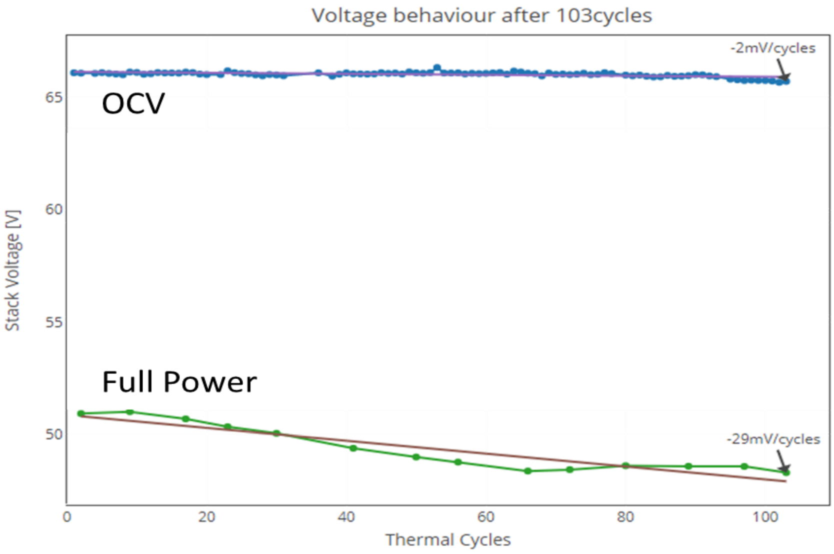


Figure 25: voltage decay after repeated thermal cycles

This result proved the improved robustness of the design B with respect to the reference case, and allowed to validate therefore the introduced design and process changes.

3.6 Stack testing

The original objectives of the work package WP7 are given here in a list.

- Stack production including QC (e.g. leak, ASR, dimensions etc.) with reference cells and improved cell design
- Short term aggressive stack testing of cells from three different production campaigns
- Long term testing of stack with baseline (i.e. reference) and improved cell designs

Delays in the stack delivery and problems encountered at AVL performing the stack tests prevented the stack tests being completed within the project.

Task 7.1 and Task 7.2 Building of stacks and short-term stack testing (SOLDpower)

After TOFC left the consortium It was decided in the project that SOLIDpower will not repeat the short-term stack tests as already carried out by TOFC.

Task 7.3 Long term stack testing (AVL, JRC)

Long term testing of stack with baseline (i.e. reference) and improved cell designs was planned. Three stack tests are planned in total: an initial reference test (meant to be a baseline test) test, a test with the first improved set of cells and a final product test in order to prove the targeted robustness. Furthermore, any new insights regarding the operation of the stack was to be considered. The goal is to prove a better robustness of the newly developed stack product compared to the stack with the reference cells. To achieve reliable results the stacks will be tested for approximately 4,000h. The data generated from these tests will be feedback into WP 5.

As a first step, a check of the testing conditions at solid power and AVL was checked and no issues were found. Furthermore, a reference cycle and harsher accelerated test cycle was agreed between AVL & Solid power. A dummy stack from solid power was used to parameterize the test stand and make sure that when the functional stack is delivered that it would be sufficiently protected with suitable shutdown procedures. Several tests were performed in order to demonstrate the functionality of the system heat up and gas supply. The operation of the reformer according to the required gas outlet composition is shown in Figure 26 left and the key temperatures within the system are shown in Figure 26 right. According to these results the test stand was deemed to be in a suitable state for testing according to the proposed test plan from WP 5 and a functional stack from solid power. A functional stack was then supplied by solid power to AVL and this stack is currently being mounted on the test bench.

Unfortunately, several issues occurred during the tests with the functional stack at AVL. These issues including power failure on the test stand and stops forced by other equipment delayed the initiation of the stack testing. After these problems were resolved it was noticed that the OCV reading on the E-load and independent voltage meter showed different values. Also, the E-load was showing a current being drawn from the stacks, but when measuring with a LEM sensor no current was measured. The values diverged even further as current was drawn from the stacks. The same issue was not observed when the E-laod was tested with batteries and it was assumed that an earthing problem was forcing the reading on the E-Load to be false. After significant fault-finding exercises no issue could be found.

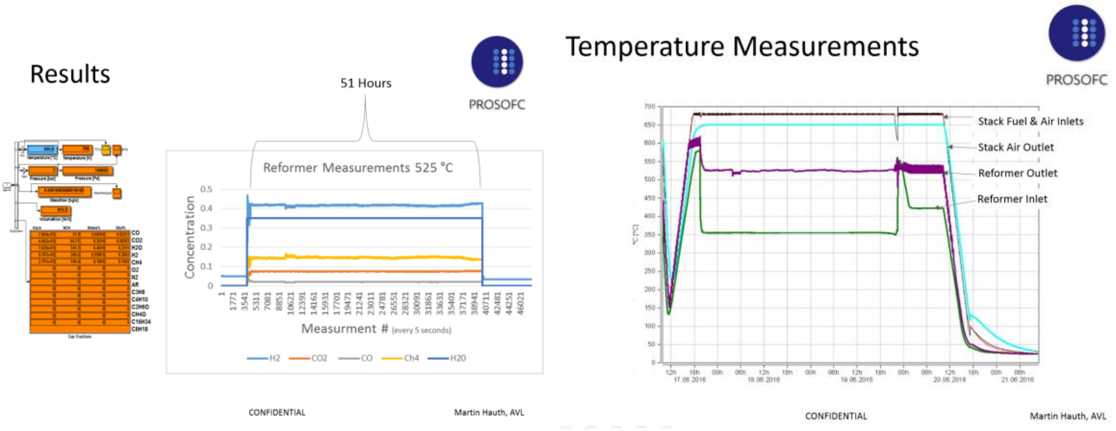


Figure 26: Example test result showing the suitable reformer operation.

After deeper analysis of the test data it was observed that when an emergency stop was activated the E-Load loaded the stack for a short period of time. A gas purge, is activated in such an event and it is believed that an event like this occurred while the stack was being supplied with a 5% H2 in 95% H2 mixture and a re-oxidation of the anode occurred. Either a single event or several events lead to a fracture in the electrolyte and the result was clearly seen during the final attempts to run the stack with a new E-Load. After this test, it was decided together with SOLIDpower to stop the stack tests. No stack tests have been carried out by JRC because the stack test rig was not appropriate for a SOLIDpower stack.

Results and achievements

The functional stack of SOLIDpower could be implemented successfully to the test stand and the test setup preparation could be finished. A proper steam reforming performance and a further conditioning of the air and fuel flows to the stack were achieved. Issues occurring during the test damaged the stack which did not allow to continue the stack test.

AVL and SOLIDpower discussed the status of the stack and decided to stop the stack test at AVL. Since at that point only little time was left in the project and no further stack was available it was decided to discontinue with stack testing. Furthermore, no stack tests have been carried out by JRC because the stack test rig was not appropriate for a SOLIDpower stack. Therefore, no stack tests could be performed to allow the originally planned validation of the stack design optimization. However, the improvements in the stack design are validated and confirmed by SOLIDpower internal stack validation program. The unused resources for stack testing at AVL were transferred to an additional task in WP4 (Task 4.7 “Optimization methodology using optiSlang based on Simulink process simulation model”). This approach was discussed and agreed on with the project officer in the course of the final project meeting in London.

3.7 Post mortem analysis & Cost analysis

The aim of WP8 is to develop tools to assess improvements of the stack technology, both in terms of technical aspects and costs. The objectives are defined in detail as follows:

- The objective of this work package is to document improvements of stack design design and processes with respect to cost and robustness of seals and cells. Thus this work package works in close cooperation with WP6 Improvement and implementation of cell design and WP7 Stack testing.
- Also, the objective of this work package is to document mechanical failures of state of art cells and seals in a stack after initialisation or operation and to supply these findings as input on cell fractures or similar mechanical failures to WP2 Continuum Material Laws and WP4 Assessment and Modelling.

Task 8.1: Post mortem analysis (HTc, EPFL)

A web-based tool has been set up that allows to create standardized post-operation reports for stacks coming back from field trials or from internal testing. The reports contain a detailed description and documentation of observations. For each stack, failure statistics are collected.

Among others:

-number of broken cells
-number of cells presenting traces of re-oxidation
-number of manifold seals that are defect
-etc. (the rest being confidential)

Not only the number of defects is recorded, but the location of the defect in the stack is tracked and used for analysis.

These numbers are then put in relation with the qualification data of the stack, that is, a set of indicators collected during the sintering and performance qualification of the stack. The database contains data about stacks having been used in systems, as well as data from production samples that are disassembled on purpose for quality monitoring.

Besides post-mortem analyses, this web-based tool contains all collected data from production and system operation, allowing for end-users to follow the evolution of quality and failures rates and causes.

By using this tool, it was possible to identify objectively typical location of failures, in particular for the fuel manifold seals. These findings were then put in relation with simulation outputs from WP4, where good agreement was found between observations and simulated predictions, hence validating the combined modeling and experimental approach. This has allowed to set priorities in terms of design and material changes for the most critical defects.

During the last year of the project, the database has shown that design changes developed in the PROSOFC project have objectively improved failure statistics, in particular by resolving one of the main failure causes found at the fuel manifold seals located in the vicinity of the stack end-plates.

Task 8.2: Cost analysis (HTc)

The cost analysis is based on the total cost of ownership (TCO) for the end-user, expressed in Euro per kilowatt-hour. This cost includes investments, maintenance, operational cost and savings on heating, if focusing on CHP applications. Therefore, the cost is a function of materials and manufacturing, but also power output, efficiency and stack lifetime.

The cost model consists therefore of three main parts: a first material and process cost model. Second, a performance and lifetime estimator. And third, a TCO calculator combining the information of the two other models.

An overview of this modeling framework is shown in the figure below.

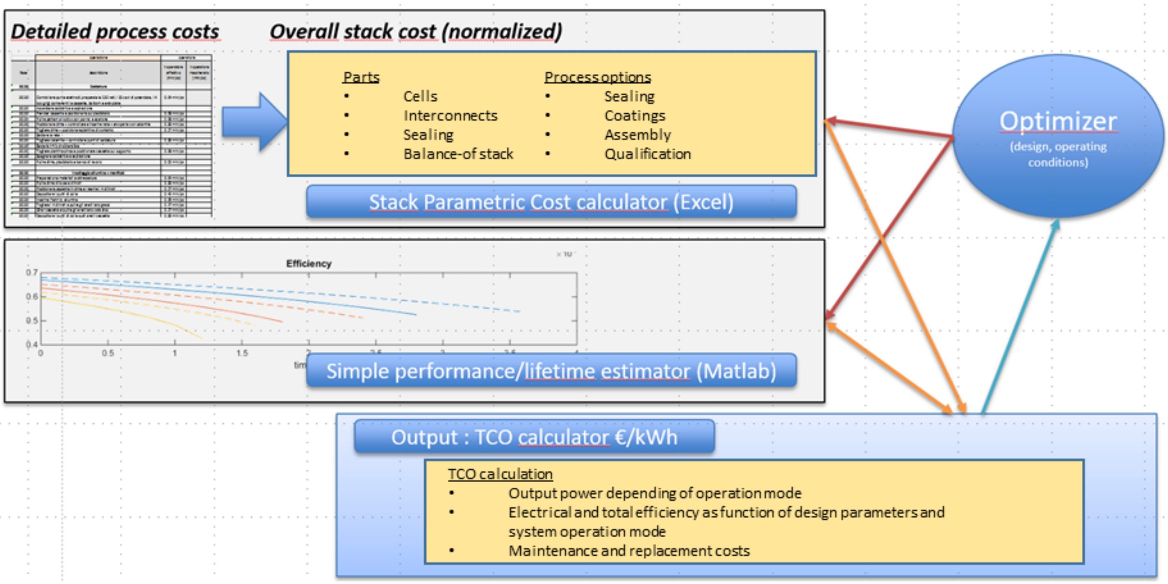


Figure 27: Cost modelling framework developed at HTceramix/Solidpower

This model is completed by the simulation of different system operating modes in a real system, in particular for the BLUEGEN system which is the main product of SOLIDpower. It is now possible to evaluate and optimize stack design and operating modes altogether. For instance, it can be assessed how far it can be beneficial to reduce the power density in the stack, both in terms of expected lifetime and in terms of TCO, or to evaluate how to deal best deal with stack ageing in system operation.

This tool is not only useful to optimize the stack and system designs for the end-user, but also to develop appropriate strategies with the marketing department.

In the figure below, an example is given for the expected stack lifetime and total energy production, for different operating modes and if varying the number of cells in the stack. It can be shown that increasing the number of cells generally improves the expected lifetimes. However, some modes (D,E) present a lower sensitivity to the number of cells, and maximize both lifetime and total energy produced. The modes cannot be described here for confidentiality reasons.

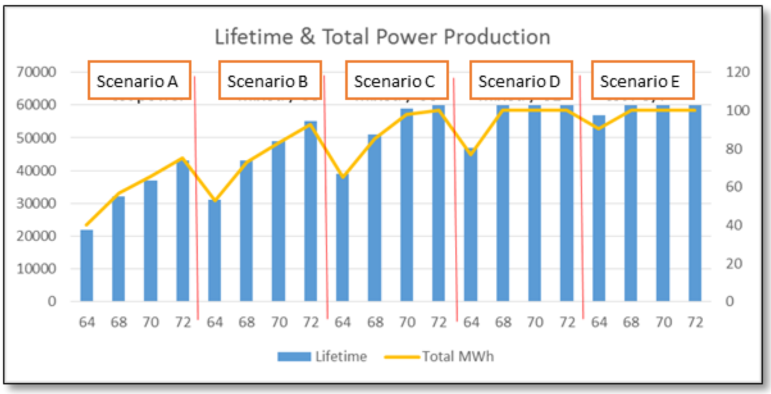


Figure 28: Stack lifetime and expected total electricity production for different operating modes and as function of the number of cells in the sack

Therefore, besides costs, this model framework now allows to evaluate other key performances indicators for the end-user and for the manufacturers, such as service intervals, system maintenance costs, product performance on the long-term, etc.

3.8 Dissemination

3.8.1 International Workshops on Mechanics in SOFCs

Two international workshops have been held in connection with the European Fuel Cell Forum (EFCF) in Lucerne, Switzerland in 2014 and in 2016. The participants came from both academia as well as the industry, with a total number of participants of 26 and 20 at the two workshops.

The overall topic of the workshops was on improving the mechanical robustness of the SOFC technology, in-line with the objective of the PROSOFC project. At the first workshop in 2014, the focus was on:

- Part I: Mechanical challenges for the SOFC technology – where should we use fracture mechanics and statistical approaches, respectively
- Part II: How to capture important phenomena in SOFC modeling with current computational power available

In 2016 the topics were:

- Part I: Brittle interfaces in SOFC stacks – the best tools to characterize and analyze them
- Part II: Flexible or rigid material components to fight the thermal stresses – guides to the engineers

Further details on the workshops can be found in D9.1 and D9.3 of the European PROSOFC project. With the completion of the two workshops, M9.1 and M9.3 were fulfilled.

Figure 29 Picture from Workshop 1 in 2014.

Figure 30 Picture from Workshop 2 in 2016.

The workshops allowed for a good dissemination of the ongoing work in PROSOFC project as well as input from external experts.

At the first workshop in 2014 in particular Edgar Lara-Curzio from Oak Ridge National Laboratory, USA gave valuable feedback to some of the mechanical testing methods applied in the project in terms of suggestions for improvements. In the workshop Briggs White, US DOE National Energy Technology Laboratory, USA (responsible for the SECA program, USA) found the collaboration between industry and academia interesting.

The second workshop in 2016 led to some interesting discussions regarding general views on stack designs. The FZJ Jülich participants (Ludger Blum, Jürgen Malzbender) mostly believed in using creep resistant materials in all components, whereas the representative from SOLIDpower, Zacharie Wuillemin, believed that some of the components of the stack should be flexible to accommodate thermal stresses (without specifying which). Generally, it can be concluded that this certainly deserves more attention from the research community.

3.8.2 Main dissemination activities

The following dissemination activities were carried our during the project duration.

Title	Date	Event	Type
FCH-JU Programme Review Day 2013	11/10/2013	FCH-JU Programme Review Day 2013	Presentation
Poster Presentation at the 11th European SOFC & SOE Forum	7/4/2014	11th European SOFC & SOE Forum	Poster
Paper Submissions to the 11th European SOFC & SOE Forum	7/4/2014	11th European SOFC & SOE Forum	Other
FCH-JU Programme Review Days 2014	11/10/2014	FCH-JU Programme Review Days 2014	Presentation
FCH-JU Programme Review Days 2015	11/18/2015	FCH-JU Programme Review Days 2015	Presentation
Presentation held at the PRIME 2016	10/11/2016	PRIME 2016	Presentation
FCH-JU Programme Review Days 2016	11/8/2016	FCH-JU Programme Review Days 2016	Presentation
Poster presentation at EFCF2016	7/5/2016	EFCF2016	Poster
Presentation held at the workshop "New Frontiers in Fuel Cell Modelling: Probabilistic Design and Open Source Platforms”	12/15/2015	Workshop "New Frontiers in Fuel Cell Modelling: Probabilistic Design and Open Source Platforms”	Presentation
Presentation held at the 14th international conference of European Ceramic Society	6/25/2015	14th international conference of European Ceramic Society	Presentation
Poster presented at the "90th DKG Annual Meeting and Symposium on High-Performance Ceramics 2015"	3/15/2016	90th DKG Annual Meeting and Symposium on High-Performance Ceramics 2015	Poster
1st invited workshop on SOFC mechanics	7/1/2014	1st invited workshop on SOFC mechanics	Workshop

2nd invited workshop on SOFC mechanics	7/5/2106	2nd invited workshop on SOFC mechanics	Workshop
FCH-JU Programme Review Days 2017	11/23/2017	FCH-JU Programme Review Days 2017	Presentation
ModVal14, 14th Symposium on Fuel Cell and Battery Modelling and Experimental Validation	2nd & 3rd of March 2017	ModVal14, 14th Symposium on Fuel Cell and Battery Modelling and Experimental Validation	Conference
2 presentations at the ECS SOFC XV	07/04/2017	ECS SOFC XV	Presentation

4 Potential impact

4.1 Exploitation of results

Exploitation of AVL foreground

AVL will use the produced foreground in the field of full-scale stack modeling, failure modeling and cell testing in the further fuel cell system development. PROSOFC significantly contributed to the further improvement of AVLs capabilities in these fields of expertise. A significant service AVL will offer to its customers will be SOFC stack modeling. The results from PROSOFC where a complete stack model based on a real stack geometry and materials was established and simulated in AVL FIRE are a valuable reference to advertise its services to the fuel cell industry. The improvements will be directly implemented into AVLs commercial CFD software product AVL FIRE. The software is sold on a license basis. Furthermore, the combination of AVL FIRE with optiSlang demonstrated a powerful method in terms of design optimization which will be further pursued not only for stack but complete system development. However, the limitations of this approach due to high processing time of the complex models will require further development to include them reasonably in a holistic simulation. This will improve the scope and quality of AVLs fuel cell engineering portfolio. In addition, the results from reliability engineering in terms of failure modeling and developing test programs to validate system reliability targets will be used in AVLs Load Matrix methodology to promote its significance for future test programs.

Exploitation of DYN foreground

DYN's business target are software sales and consulting work on high-level engineering projects. This implies an audience in many engineering fields. DYN therefore does not follow specific plans for SOFC community. During the PROSOFC project it turned out that SOFC engineers use a large number of commonly available simulation tools (e.g. COMSOL, MATLAB, g-PROMS, NASTRAN, ABAQUS, ANSYS). DYN's approach is to create a software that can interact with all of these tools. This lead to the development of optiSLang and SoS.

Exploitation will focus on customers/users of the above mentioned CAE solvers (e.g. ANSYS and/or ABAQUS). PROSOFC provided an excellent opportunity to demonstrate the usefulness of DYN's software in a highly interdisciplinary context such as the analysis of SOFC. Current sales channels (ANSYS International, CADFEM (D-A-CH and International), DYNARDO U.S. Inc. (to be founded in 2018) will be used to further propagate licensing of optiSLang and SoS. Results from PROSOCF will be presented at international industry-oriented conferences (e.g. CASCON and WOST conferences in 2018).

The expected impact is on one hand related to improvements of software sales activity of optiSLang (in all engineering areas), and – more importantly – related to the development of new areas of application of DYNARDO software (e.g. modelling if contact layer variations, analysis of measurements, meta modelling of field data).

Already as of now, DYN has been able to acquire new projects using the methodology developed within PROSOFC in different fields: Automotive engineering (e.g. DAIMLER); Aircraft engineering; Power generation (e.g. SIEMENS); Suppliers (e.g. BOSCH); Biomechanics (e.g. research at UK Aachen).

Further research/work will be necessary. The main points are: Analysis of transient models, of measurements with large time signals or of many samples; more accurate meta models together with improved sensitivity measures; further improved usability of the software.

Exploitation of DTU foreground

To analyze the mechanical behavior of SOFC stack components at elevated temperatures and in controlled atmosphere, DTU developed new experimental methods (to characterize transient creep) and new modelling approaches (homogenization for effective SOFC stack modelling).

The foreground will primarily be used to assist SOFC stack manufacturers to analyze their stacks and mitigate possible mechanical challenges. The aim is to do this within European as well as national funded projects.

With a role as knowledge provider rather technology provider, DTU has primarily focused on generating publicly accessible knowledge, and not generated any IPR.

Towards the project closure, a new creep phenomenon was observed using the SOLIDpower steels. The consequence of this could be important to understand for the integrity of their stacks. Also, good models for to predict the failure of sealings are yet to be developed. This should clearly be part of future research, given the challenges of sealing failures (for many SOFC stack manufacturers).

The scientific findings throughout the project are published in reputable international scientific journals, and have already attracted some attention. This will for sure impact the development of the SOFC technology.

Exploitation of KIT foreground

The KIT “Institute of Applied Materials – Electrical and Electronic Engineering”, is focusing on the electrochemical and microstructural characterization and modeling of fuel cells and batteries. Testing methodologies and models are continually developed in national and European research projects. In PROSOFC new experiences were gathered in testing of two types of anode supported cells. Furthermore approaches to reconstruct the microstructure of complex multiphase electrodes by FIB/SEM tomography refined.

The foreground of PROSOFC is already exploited in ongoing research projects as well as in the education of our students at KIT.

As a university institute we are aiming at the dissemination of our results in lectures and papers, therefore no IPR exploitable measures are intended.

Electrochemical testing methodologies and models will continuously be developed and applied in different research projects.

4.2 Main dissemination activities

The following table gives an overview over the dissemination activities performed by the project consortium throughout the project.

Table 1: Main Dissemination Activities

Title	**Date**	**Event**	**Type**
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5 General Project Information

Project Acronym	PROSOFC
Project Long Name	Production and Reliability Oriented SOFC Cell and Stack Design
Call topic	FCH-JU-2012-1
Start-date	01th May 2013
End-date	31st October 2017
Total budget	€ 7,331,214.71
FCH JU contribution	€ 3,011,000.00

Project Consortium:

Partner	Country
AVL List GmbH	Austria
Dynardo Austria GmbH	Austria
Danmarks Tekniske Universitet	Denmark
Forschungszentrum Jülich	Germany
Karlsruher Institut fuer Technologie	Germany
Imperial College of Science, Technology and Medicine	United Kingdom
JRC – Joint Research Centre – European Commission	Belgium
HTceramix S.A.	Switzerland
EPFL - École polytechnique fédérale de Lausanne	Switzerland

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