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Project: **“Solid Oxide Fuel Cells for the Renewable Energy Transition”**

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1. Project details

Project title	Solid Oxide Fuel Cells for the Renewable Energy Transition
Project identification	Energinet.dk project no. 2014-1-12231 (ForskEL)
Project period (date, year)	01.08.2014 – 31.03.2017
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2. Executive summary

Wind and solar power are rapidly dominating new electricity generation installations around the world, and Denmark is at the forefront of this transition. In 2016, the fraction of *new* power generation that came from renewables was more than half globally, nearly 90% in Europe, and in Denmark already more than half of the *total* generation capacity was renewable¹. Since wind and solar sources only produce power when the wind blows and sun shines, technological solutions to manage these intermittent supplies are urgently needed. This project investigated the use of solid oxide electrochemical cells (SOCs) to balance the future grid. When operated as fuel cells (SOFCs), they produce electricity and heat from fuels like natural gas and biogas with higher electrical efficiencies than conventional power plants and at a smaller, distributed scale. SOFCs can provide dispatchable power generation to fill in the gaps of wind/solar supply.

A unique ability of SOC technology compared with conventional power generation is that SOCs can also be run in reverse, in electrolysis mode, to store excess renewable electricity as chemical fuels with energy efficiencies approaching 100% depending on the fuel. The possible fuels include liquid hydrocarbons like diesel, which can be used in the transportation sector, or methane, which can be injected into the gas grid and used later for power generation like a giant battery². This reversibility is an advantageous capability of SOCs and was an important focus point in this project.

The project built on the results of previous ForskEL projects that had helped develop SOC technology for fuel cell applications (Towards Smart Grid Ready SOFC, Energinet.dk project no. 2012-1-10747) and electrolysis applications (Development of SOEC Cells and Stacks, Energinet.dk project no. 2011-1-10609). Despite having been developed to a high level of performance, SOCs were still not widely commercialized. Comprehensive testing in the laboratory, as well as field testing, points to a big challenge: attaining highly stable device performance under real conditions over many thousands of hours. Device lifetime is a key factor in the cost of SOC technology. Accordingly, this project first

¹ UN report, *Global Trends in Renewable Energy Investment*, 2017; [Article in The Guardian](#), 9 Feb. 2017; and Danish Energy Agency

² Graves et al, *Sustainable hydrocarbon fuels by recycling CO₂ and H₂O with renewable or nuclear energy*, *Renew. and Sust. Energy Rev.* 15 (2011) 1–23.
Graves et al, *Eliminating degradation in solid oxide electrochemical cells by reversible operation*, *Nat Mater.* 14 (2015) 239–244.
Jensen et al, *Large-scale electricity storage utilizing reversible solid oxide cells combined with underground storage of CO₂ and CH₄*, *Energy Environ. Sci.* 8 (2015) 2471–2479.

focused on identifying and understanding the most prevalent lifetime limiting factors and used this knowledge for further development of long-living devices.

Work-package 1 (WP1) therefore started with a comprehensive review of lifetime-limiting factors based on more than 100 publications and corresponding 650 000 hours of testing of cells and stacks, as well as 180 single cell tests carried out in the past at DTU Energy, obtaining a quantitative analysis of the current international status of degradation and lifetime in the field [1-4]. One important observation was that test lengths well in excess of the usual 1000 h are necessary to establish meaningful degradation rates, and therefore tests longer than 2 years were run in WP2. Another key finding was that the nickel and yttria-stabilized zirconia (Ni-YSZ) fuel-electrode is responsible for most modes of degradation, as well as hard failure mechanisms such as cell destruction by nickel oxidation and carbon deposition. This motivated the cell development activities in WP3 to focus on replacing the nickel-based fuel-electrode with a more stable material. Besides performance degradation of device materials, the high operating temperature of SOCs can also, with careful control, lead to improvement of the properties of materials. A review of such re-activation mechanisms was also undertaken [5] and used in designing experimental investigations of performance regeneration in WP3.

Besides identification of degradation mechanisms and trends in degradation rates, it is necessary to determine market applications and the level of degradation that may be acceptable. The second part of WP1 therefore entailed a techno-economic analysis of a new market scenario utilizing a single device in reversible operation mode – buying electricity and selling methane fuel product, or vice versa, when the market price spread was sufficient to obtain revenue. This analysis used realistic performance and durability data, system design, operating profiles, and Denmark grid market data. Using presently available SOC technology and today's market prices, the system was not profitable in this arbitrage-like scenario. However, analysis of future scenarios – with a higher fraction of wind power and resulting increase in the price spread, and with SOC technology improvements – showed profitability [6].

Work-package 2 (WP2) dealt with long-term testing, which in addition to operation of cells for thousands of hours also involved development of advanced methods of testing and analysis. Simplified electrodes were used with advanced electrochemical and spectroscopic techniques to investigate electrode kinetics and degradation phenomena, enabling isolation of the response of the single component which is inaccessible in measurements made on the full, complex device [7-9,25,24]. Detailed impedance spectroscopy measurements on the actual, full cells fed with realistic fuels were used to link the component measurements to the full-cell geometry [10-12]. The measurements were then used in multi-physics models of the full device that were constructed to determine in 3D with a new level of accuracy where local “hotspots” of degradation can occur and provide input for improvement of cell design [23,26-27].

Full cells were operated at constant conditions at 700 °C for more than 2 and 1 years with hydrogen and internally-steam-reformed methane fuel supplies, respectively. These tests used clean nickel and gold contact components on the fuel and air sides, respectively, and provided baseline stability data with degradation rates as low as 2.5 mV per 1000 h (0.3% voltage drop) during 1 year of operation, accelerating degradation during the test, and faster degradation for the methane test. These low degradation rates are commercially promising, however actual devices with steel stack components and with dynamic operation profiles might lead to higher degradation. Stack-element testing using steel contact components was also carried out, both for comparison as well as for accelerated testing. During a 1000 h test, the degradation profile followed that of the baseline test remarkably, indicating that the steel components did not cause any increased degradation during this period, although the test conditions were slightly different. Steam content in the fuel gas was used as an acceleration vector and showed a strong effect on degradation of “fresh” Ni-YSZ fuel-electrodes (during the initial testing period) but little effect on an already-aged electrode. Long-term dynamic operation in reversible mode (periodic switching between fuel-cell and electrolysis modes) revealed that gaseous impurities cause fast degradation, which does not occur in steady-state fuel-cell mode. With a

cleaned fuel gas supply, operating in reversible mode showed much lower degradation than in the steady-state fuel-cell mode without gas cleaning [12].

Work-package 3 investigated ways to improve lifetime and efficiency by: *in-situ* repair/regeneration during operation, *ex-situ* repair/regeneration, and development of new and improved cells. *In-situ* regeneration is a new concept that entails careful adjustment of operating conditions (gas composition or applied voltage) to regain performance, usually of electrodes [5]. We tested high cathodic polarization of Ni-YSZ electrodes [29] as well as oxidation/reduction cycles of fuel-electrodes [13,28], both of which were able to restore performance by regaining reaction site density. Limitations of these techniques were also considered. *Ex-situ* regeneration involved cooling down the device, infiltrating nanoparticles in the fuel-electrode and heating back up, and was also shown to restore performance [2,30].

Finally, in WP3 completely new cells were developed that did not contain nickel in the fuel-electrode. By removing nickel, the source of many modes of degradation, a cell with more robust electrodes was obtained. First, new fuel-electrodes comprised mainly of nanostructured Gd-doped CeO₂ were developed [7,13-15,28,31,32]. They provided a polarization resistance as low as 0.01 Ω cm² at 600 °C in hydrogen/steam fuel environment, an order of magnitude lower than conventional nickel-based electrodes, and they were stable against carbon deposition and oxidation. For practical porous electrodes, Gd-doped CeO₂ provides insufficient electronic conductivity so a small fraction of SrFe_{0.75}Mo_{0.25}O₃ was added. Since the Ni-YSZ is the usual support-layer for thin-electrolyte cells and the goal was to remove nickel, oxygen-electrode-supported cells were developed and therefore stable and high-performance oxygen-electrodes that do not contain cobalt, with Pr-doped CeO₂ (PDC) as an electrocatalytic material, were also developed. In fact, PDC was used in both fuel and oxygen electrodes [14]. Full 5x5 cm² cells made with these fuel and oxygen-electrodes showed equivalent electrode performance and higher stability than state-of-the-art Ni-YSZ supported cells. Carbon deposition and oxidation treatments caused no damage, and actually the latter improved the fuel-electrode performance, suggesting that these new cells could be a game-changer for SOC technology commercialization. For future scale-up purposes, electrode backbone structures made by freeze-casting were also developed [16].

Overall the project results:

- provided a systematic, quantitative overview of device lifetime reports;
- investigated a new market application with reversible operation;
- measured the long-term stability of the devices and individual components in realistic and dynamic operation, and established new testing and analysis methods;
- developed novel ways to achieve improved lifetime by performance-regeneration treatments and new cells comprised of high-performance, more stable materials.

Further development of the latter especially is expected to be a highly promising path forward towards commercialization of the technology for all envisioned applications.

Dissemination of the project outcome has resulted in 24 published journal articles and PhD/MSc theses, 8 articles that are in “submitted” or “in preparation” state and are expected to be published soon, and 11 presentations at international conferences.

3. Project results

3.1. Overview and milestones

The project was organized into three main technical work-packages (WPs). In WP1, a comprehensive, quantitative review of reported degradation rates and lifetime-limiting factors of SOCs was carried out. The results were used to design long-term tests and accelerated tests in WP2 and to motivate development of new cells and regeneration treatments in WP3. WP1 also included a techno-economic analysis involving simulation of reversible SOC operation using real time-series electricity and gas supply and demand market pricing.

In WP2, new methods of testing and analysis for individual components of SOC devices were developed and multi-physics models were constructed to gain insight into local conditions that lead to degradation. Very long-term cell tests (longer than 1-2 years) were carried out to gain insight into long-term trends. Accelerated and dynamic tests were also done and compared with steady-state tests.

In WP3, novel treatments were developed to regenerate cell performance after cells had experienced degradation, with focus on regenerating performance of the nickel-based fuel-electrode, which is the main site of performance loss. Completely new cells were developed that did not contain any nickel yet matched the performance of the state-of-the-art nickel-based cells and showed higher stability.

The project structure is as follows:

- WP1 – Analysis of SOFC lifetime for stationary applications
 - WT1.1 – Review lifetime limiting mechanisms
 - WT1.2 – Analysis and modelling of lifetime and energy system applications
- WP2 – Durability studies
 - WT2.1 – Development of advanced methods for testing and analysis
 - WT2.2 – Realistic operation
 - WT2.3 – Accelerated testing
- WP3 – Improving lifetime and efficiency
 - WT3.1 – In-situ repair/regeneration to extend lifetime
 - WT3.2 – Ex-situ repair/regeneration to extend lifetime
 - WT3.3 – Improved cells

The project milestones and their status at the completion of the project were as follows:

M1.1: Identification of the most important reported lifetime-limiting factors & proposed test methods
Milestone fulfilled. Two comprehensive reviews [1-4] were completed, which summarized and quantified reported lifetime-limiting factors based on more than 100 publications and corresponding 650 000 hours of testing of cells and stacks, as well as 180 single cell tests carried out in the past at DTU Energy. A quantitative analysis of the current international status of degradation and lifetime in the field was obtained. The key findings were used as the basis for activities in WP2 and WP3.

M1.2: Parametric techno-economic analysis based on realistic performance and durability data, system design, operating profiles, and grid market data
Milestone fulfilled. The techno-economic analysis focused on a new market application of reversible operation, where a single reversible SOC system was designed and simulated using re-

al time-series market prices in Denmark to buy low-cost electricity and sell methane produced by electrolysis, or vice versa buying natural gas and selling electricity [6].

M2.1: Implementation of accelerated lifetime tests

Milestone fulfilled. In order to investigate realistic cell-contacting and stack related impacts on performance stability, a test set-up for long-term testing was developed and validated that used a realistic material set – steel interconnect contact/flow components – instead of the cleaner contact components made of nickel and gold used in the ideal single-cell set-up.

M2.2: Improved analysis methodology developed and demonstrated on experimental data

Milestone fulfilled. Using simplified cells, electrochemical measurements and X-ray photoelectron spectroscopy measurements were obtained to investigate detailed electrode kinetics and degradation phenomena at the isolated electrode interfaces, which are inaccessible in measurements made on the full, complex device [7-9,25,24]. The obtained electrode kinetics data was used, together with detailed impedance spectroscopy measurements on full cells [10-12], in 3D multi-physics models to determine where local “hotspots” of degradation can occur and provide input for improvement of cell design [23,26-27].

M2.3: Baseline cell and stack-element/short-stack tests in realistic operating profiles completed

Milestone fulfilled. Full cells were operated at constant conditions at 700 °C for more than 2 and 1 years with hydrogen and internally-steam-reformed methane fuel supplies, respectively. Long-term (nearly 2000 h) stack-element testing using steel contact components was also carried out, both for comparison as well as for accelerated testing.

M2.4: At least one accelerated component test and one accelerated stack-element/short-stack lifetime test completed

Milestone fulfilled. Stack-element testing was carried out for more than 3000 h to investigate the accelerated degradation effects of steam content in the fuel gas. Ni-YSZ electrode component testing was carried out using half-cells to determine the impact of initial electrode structure and initial NiO reduction temperature on long-term Ni particle size and electrode structure.

M2.5: Two different types of dynamic tests clearly showing beneficial or detrimental effects on the degradation rate compared with steady-state reference tests

Milestone fulfilled. The impacts of cyclic reversible operation (switching between fuel-cell mode and electrolysis mode) and cleaning the inlet gasses were investigated in a single-cell test series. Cyclic operation with non-cleaned inlet gasses was detrimental and cyclic operation with cleaned inlet gasses was beneficial to long-term stability.

M3.1: Assessment of the improvement levels and limitations (including repeatability) of at least two methods to counteract performance loss by adjustment of operating parameters

Milestone fulfilled. Two *in situ* performance regeneration methods were developed: high cathodic polarization of Ni-YSZ fuel-electrodes [29] and oxidation/reduction cycles of cerium oxide based fuel-electrodes [13,28], both of which modified the electrode interfaces and regained reaction site density.

M3.2: Demonstration of an improved method for ex-situ lifetime enhancement compared with the previous initial reports

Milestone fulfilled. *Ex-situ* regeneration was demonstrated by cooling down a degraded cell, infiltrating nanoparticles in the Ni-YSZ fuel-electrode and heating back up [2,30]. A previous initial report about this method did not show improvement in fuel-electrode performance as shown in this work.

M3.3: Detailed characterization and long-term stability testing under steady-state and dynamic conditions of a cell with improved initial performance or long-term stability

Milestone fulfilled. Full 5x5 cm² oxygen-electrode-supported cells with new nanostructured doped-ceria electrocatalysts in both fuel- and oxygen-electrodes were developed and tested. They showed equivalent electrode performance and higher stability than state-of-the-art Ni-YSZ supported cells. Carbon deposition and oxidation treatments caused no damage, and actually the latter improved the fuel-electrode performance.

The project was carried out according to the following timeline:

	Q1	Q2	Q3	Q4	Q5	Q6	Q7	Q8	Q9	Q10	Q11
WP1 – Analysis of SOFC lifetime for stationary applications											
WT1.1 – Review lifetime limiting mechanisms	█	█	█	█							
M1.1: Identification of the most important reported lifetime-limiting factors and proposed test methods			█								
WT1.2 – Analysis and modelling of lifetime and energy system applications		█	█	█	█	█	█	█			
M1.2: Parametric techno-economic analysis based on realistic performance and durability data, system design, operating profiles, and grid market data								█			
WP2 – Durability studies											
WT2.1 – Development of advanced methods for testing and analysis	█	█	█	█	█	█	█	█	█		
M2.1: Implementation of accelerated lifetime tests								█			
M2.2: Improved analysis methodology developed and demonstrated on experimental data							█				
WT2.2 – Realistic operation		█	█	█	█	█	█	█	█	█	
M2.3: Baseline cell and stack-element/short-stack tests in realistic operating profiles completed						█					
WT2.3 – Accelerated testing		█	█	█	█	█	█	█	█	█	
M2.4: At least one accelerated component test and one accelerated stack-element/short-stack lifetime test completed										█	
M2.5: Two different types of dynamic tests clearly showing beneficial or detrimental effects on the degradation rate compared with steady-state reference tests				█							
WP3 – Improving lifetime and efficiency											
WT3.1 – In-situ repair/regeneration to extend lifetime			█	█	█	█	█	█	█	█	█
M3.1: Assessment of the improvement levels and limitations (including repeatability) of at least two methods to counteract performance loss by adjustment of operating parameters											█
WT3.2 – Ex-situ repair/regeneration to extend lifetime			█	█	█	█	█	█	█	█	█
M3.2: Demonstration of an improved method for ex-situ lifetime enhancement compared with the previous initial reports											█
WT3.3 – Improved cells	█	█	█	█	█	█	█	█	█	█	█
M3.3: Detailed characterization and long-term stability testing under steady-state and dynamic conditions of a cell with improved initial performance or long-term stability											█

3.2. WP1 – Analysis of SOFC lifetime for stationary applications

WT1.1 – Review lifetime limiting mechanisms

This work-package started with a comprehensive review of lifetime-limiting factors based on more than 100 publications and corresponding 650 000 hours of testing of cells and stacks, as well as 180 single cell tests carried out in the past at DTU Energy, obtaining a quantitative analysis of the current international status of degradation and lifetime in the field [1-4]. Although direct comparison between long-term tests reported by different research groups and companies was challenging due to lack of standardized reporting of test conditions and results, some key trends were found. The data reported in the field overall showed a general trend of lower degradation for more recent reported tests (Figure 1). One important observation was that test lengths well in excess of the usual 1000 h are necessary to establish meaningful degradation rates (Figure 2), and therefore tests longer than 2 years were run in WP2.

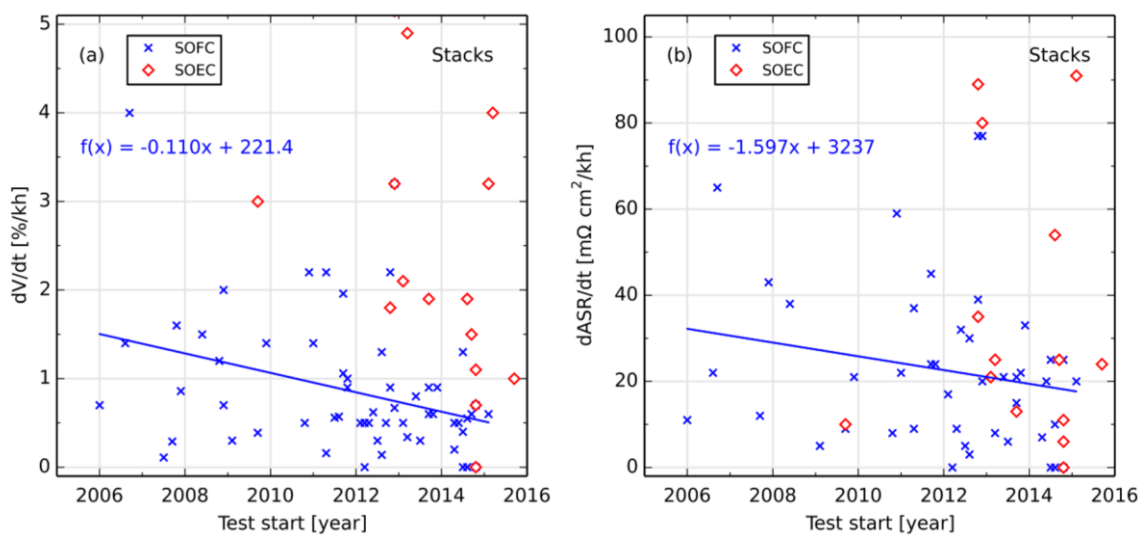


Figure 1. Degradation rate in (a) V%/kh and (b) $m\Omega\text{ cm}^2/\text{kh}$ vs the date the stack test was started (estimated by publication year and test length). The legend indicates the operation mode. From [1].

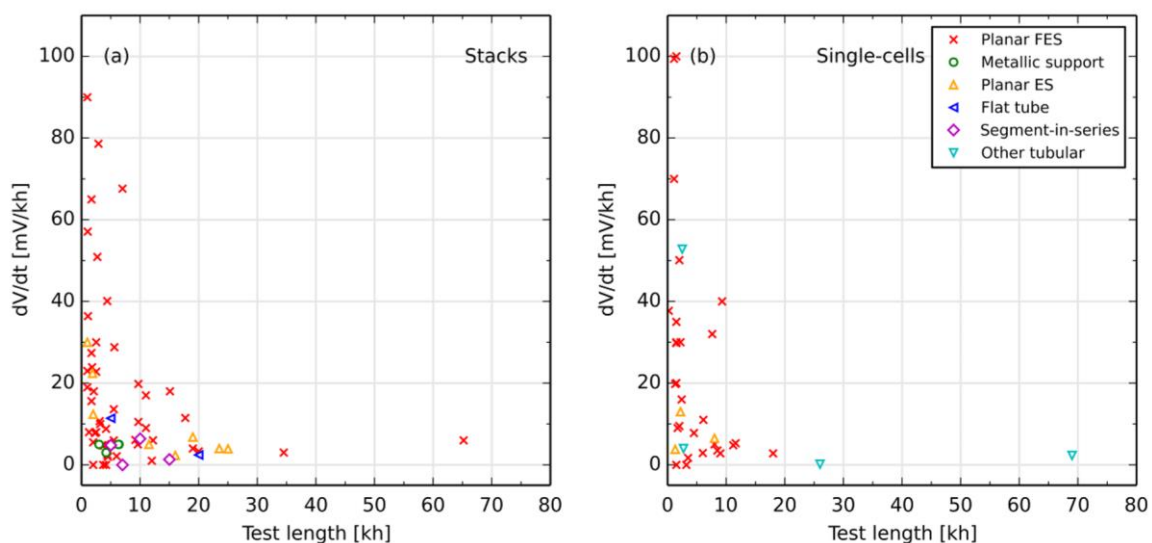


Figure 2. Degradation rate in mV/kh vs length of the reported (a) stack- or (b) single-cell-test. The legend indicates the design of the cells, where FES stands for fuel electrode supported and ES for electrolyte supported. From [1].

Another key finding was that the nickel and yttria-stabilized zirconia (Ni-YSZ) fuel-electrode is responsible for most modes of degradation, as well as hard failure mechanisms such as cell destruction by nickel oxidation and carbon deposition. This motivated the cell development activities in WP3 to focus on replacing the nickel-based fuel-electrode with a more stable material. Besides performance degradation of device materials, the high operating temperature of SOCs can also, with careful control, lead to improvement of the properties of materials³. A review of such re-activation mechanisms was also undertaken (Figure 3) [5] and used in designing experimental investigations of performance regeneration in WP3.

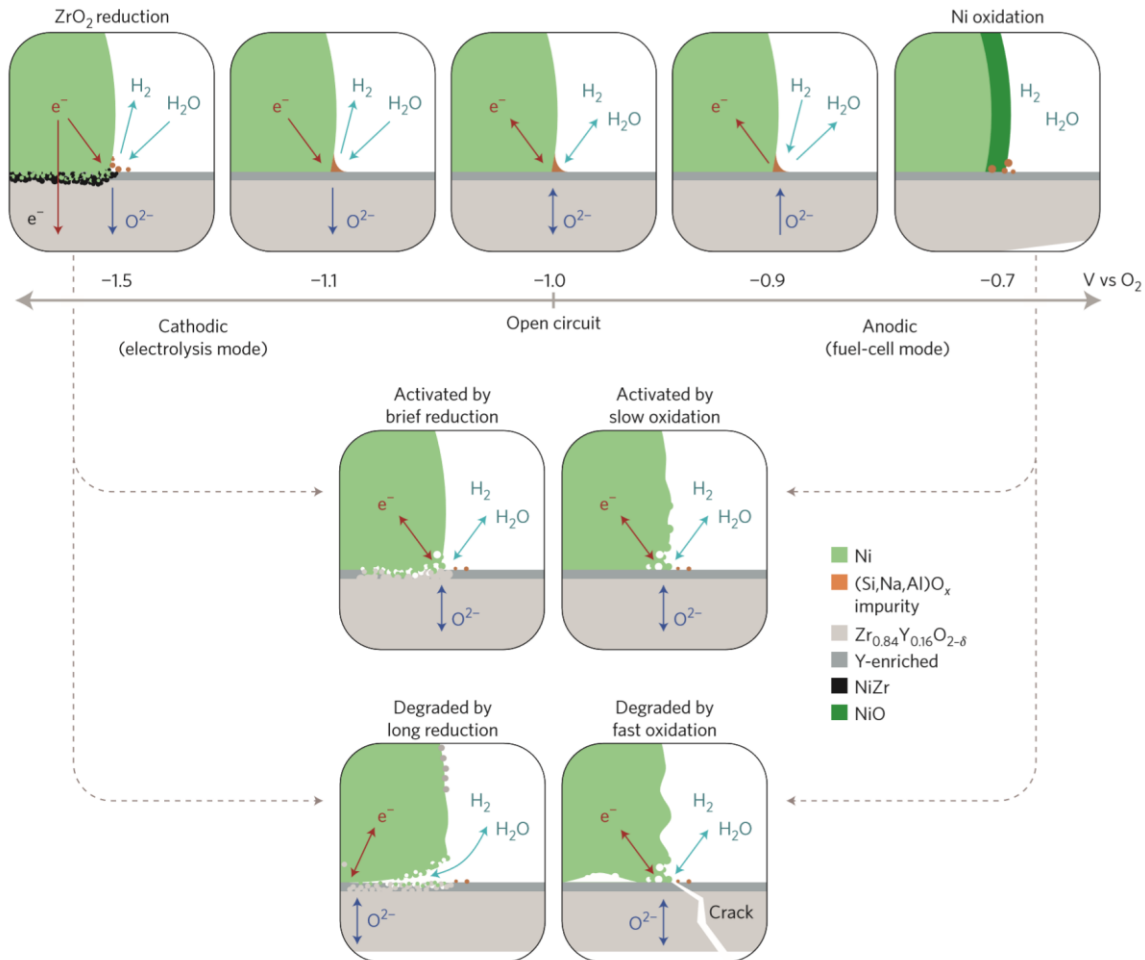


Figure 3. Ni-YSZ fuel-electrode potential-driven activation and passivation phenomena. From [5]. The cited article also illustrates similar oxygen-electrode activation and degradation phenomena.

The most widely reported lifetime-limiting factors can be summarized as follows. The affected locations in the solid oxide cell where the degradation modes occur are labeled in Figure 4 with the corresponding numbers in the following list.

Electrode degradation mechanisms – intrinsic / difficult to avoid:

1. Detrimental structural evolution of the fuel-electrode in the form of loss of percolation of nickel particles and coarsening of nickel particles (leading to loss of electronic conduction and reaction site concentration)

³ C. Graves, *Reversing and Repairing Microstructure Degradation in Solid Oxide Cells during Operation*, ECS Trans. 57 (2013) 3127–3136.

2. Poisoning of the oxygen-electrode by chromium vapor species from stainless steel interconnects and other system components
3. Deactivation of electrode reaction sites by segregation of elements from the material bulk to its surface

Mechanical failure mechanisms:

4. Loss of cell/interconnect contact due to creep
5. Delamination of cell layers due to differing thermal expansion and other stresses induced by dynamic operation
6. Rapid damage from an uncontrolled Ni-NiO oxidation-reduction cycle or carbon deposition on nickel from carbonaceous fuel gasses supplied without enough steam

Electrode degradation by gaseous impurities – extrinsic / avoidable:

7. Poisoning of the fuel-electrode by impurities, e.g. adsorption of sulfur and phosphine from the gas supply on nickel
8. Poisoning of the oxygen-electrode by trace impurities in air like sulfur dioxide

Other issues:

9. Resistivity increase between cell layers due to solid-solid interfacial reactions
10. Issues arising from insufficient sealing

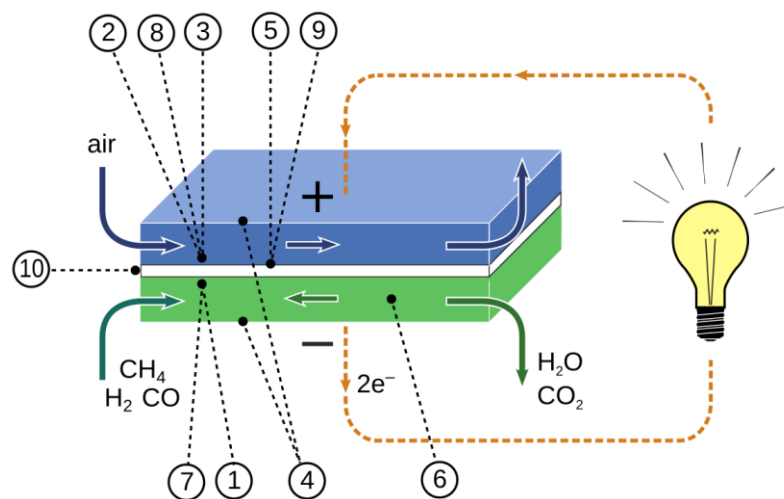


Figure 4. Diagram of a solid oxide cell in fuel-cell mode with the locations of the most important degradation mechanisms labeled.

Some of these factors can be addressed by system design and others by cell materials design, and it should be possible to test them in a compressed way (without requiring a set of several-year-long tests). For example, considering the first two degradation mechanisms in the list: (1) Ni mobility is known to increase with temperature, current and steam partial pressure, and therefore those test parameters are proposed as accelerating vectors for Ni particle coarsening and resulting performance degradation. (2) The concentration of Cr vapor species is known to be higher at higher temperature, indicating also that temperature is also a possible acceleration parameter for that Cr poisoning of the oxygen-electrode. Further work will include determining how to try to isolate such acceleration parameters in such a way that not too many degradation modes are accelerated simultaneously – e.g. if Ni mobility and Cr poisoning are the only two accelerated mechanisms then it should be possible to study both with careful testing and analysis, but additional simultaneous mechanisms will likely make it impossible to determine their individual impacts.

WT1.2 – Analysis and modelling of lifetime and energy system applications

Besides identification of degradation mechanisms and trends in degradation rates, it is necessary to determine market applications and the level of degradation that may be acceptable. The second part of WP1 therefore entailed a techno-economic analysis of a new market scenario utilizing a single device in reversible operation mode – buying electricity and selling methane fuel product, or vice versa, when the market price spread was sufficient to obtain revenue. This analysis used realistic performance and durability data, system design, operating profiles, and Denmark grid market data.

First, good estimates of the capital cost of the stack and system are necessary. The stack cost scales with production volume, as with all mass-produced technology. It was found that all of the stack cost estimates in literature agree rather well and approach ~\$800/m² or \$100-200/kW at large production volume (Figure 5).

A generic system schematic is shown in Figure 6, where each “conditioning” block is comprised of several system components such as heat exchangers, compressors, etc. In literature, system costs are typically a factor of 5 to 10 times that of the stack due to all of these additional components. The system cost has been carried out with traditional chemical plant scaling laws, which show the well-known trend of lower cost per kW with larger plant size.

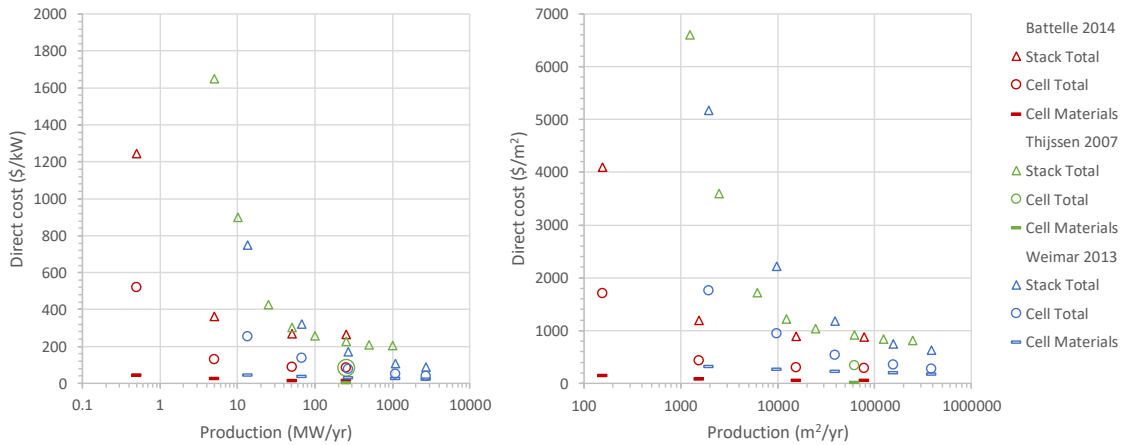


Figure 5. Comparison of literature estimates of stack capital cost vs mass-production volume.

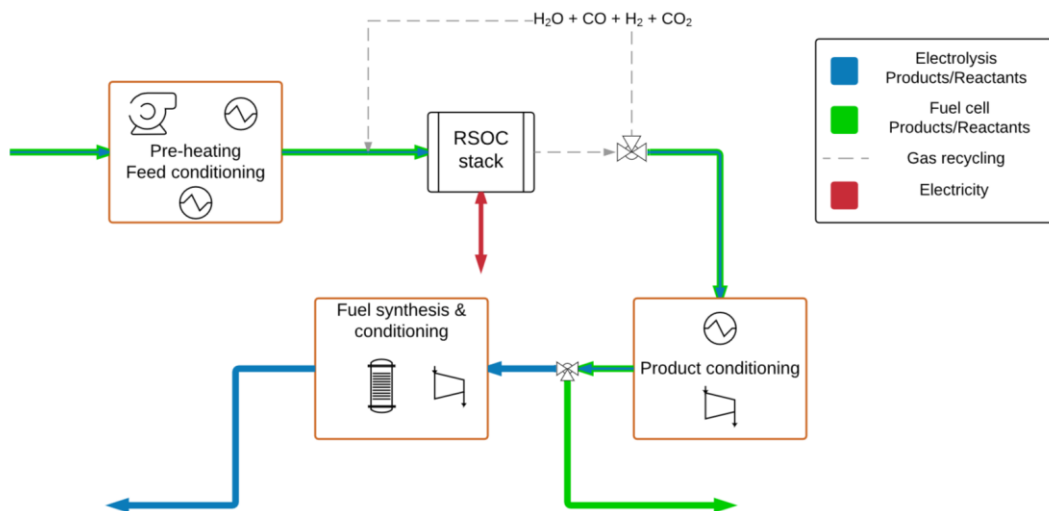


Figure 6. Generic “open-ended” reversible SOC system. From [6].

A model has been developed that finds the optimal operating point at each point in time, using time-series market prices for electricity in natural gas in Denmark to perform the optimization. The solid oxide cell stack and system are reversible and can operate either in fuel-cell mode or electrolysis mode, producing electricity from natural gas or producing methane from electricity, depending on the price signals. This represents a scenario where such a system would be installed today at a small scale such that it would not influence the electricity and gas markets. The goal is to determine how the system can be designed and operated most profitably in current and future markets.

The simulation has been run using a number of different scenarios that varied the market price data (recent prices as obtained from Energinet.dk, and future predicted prices from different studies) as well as system characteristics such as stack resistance, limits on the stack operating current or voltage, and degradation rate. Results for recent prices and a base-case system characteristics scenario, representing realistic parameter values based on today's available technology and system design, are shown in Figure 7. The spot market electricity price swings enable bursts of considerable revenue by buying low-cost electricity and operating in electrolysis mode to produce and sell methane with the system. Furthermore, when looking at each year of time-series data, the fraction of time spent in fuel-cell mode operation decreases and the fraction of electrolysis mode operation increases.

The system profitability is greater for the reversible system than for either a fuel cell or electrolysis system, even when taking into account the slightly higher system cost in a reversible system. Using presently available SOC technology and today's market prices, the system was not profitable in this arbitrage-like scenario. However, analysis of future scenarios – with a higher fraction of wind power and resulting increase in the price spread, and with SOC technology improvements – showed profitability [6].

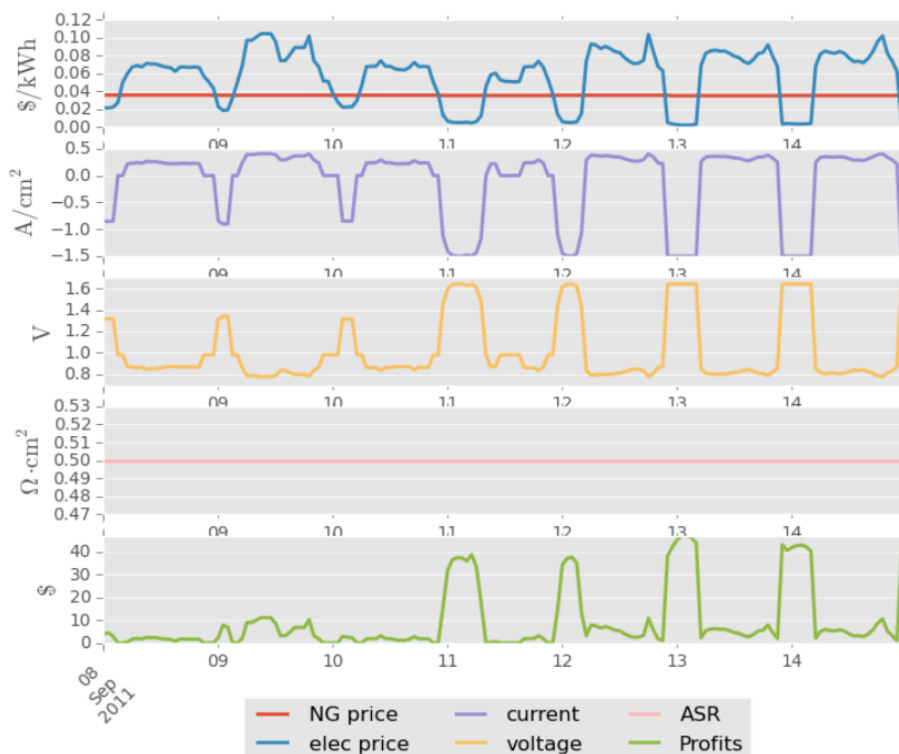


Figure 7. Simulation of reversible SOC operation using one week of spot market time-series data. From [6].

3.3. WP2 – Durability studies

This work-package comprised systematic testing of state-of-the-art SOCs produced at DTU Energy on component level and in single-cell configuration, with the overall goal to further the knowledge of performance stability in (i) realistic operating conditions and (ii) to find the limits of performance stability in accelerated treatments.

WT2.1 – Development of advanced methods for testing and analysis

In addition to testing cells for thousands of hours, this WP also involved development of advanced methods of testing and analysis.

For designing accelerated lifetime testing methods in a simulated stack environment, the single-repeating-unit (SRU) or stack-element methodology was applied. Figure 8 shows the test setup that has been developed for SRU testing. The cells were placed in between Cofer22 APU interconnect plates with etched gas distribution channels, and glass seals were used. The gas was supplied via counter-flow geometry. On the fuel-electrode side, the interconnect plate was used without any protection layer while on the oxygen electrode side a 70-80 μm thick manganese cobalt ferrite spinel protective layer was applied via atmospheric plasma spraying. The testing procedure started by heating the single repeating unit up to 900°C for sealing and the NiO of the contact layer, support and fuel electrode was reduced for 2 h in a 9:91 $\text{H}_2:\text{N}_2$ gas mixture at 20 l h^{-1} and completed for 1 h in pure H_2 at 20 l h^{-1} .

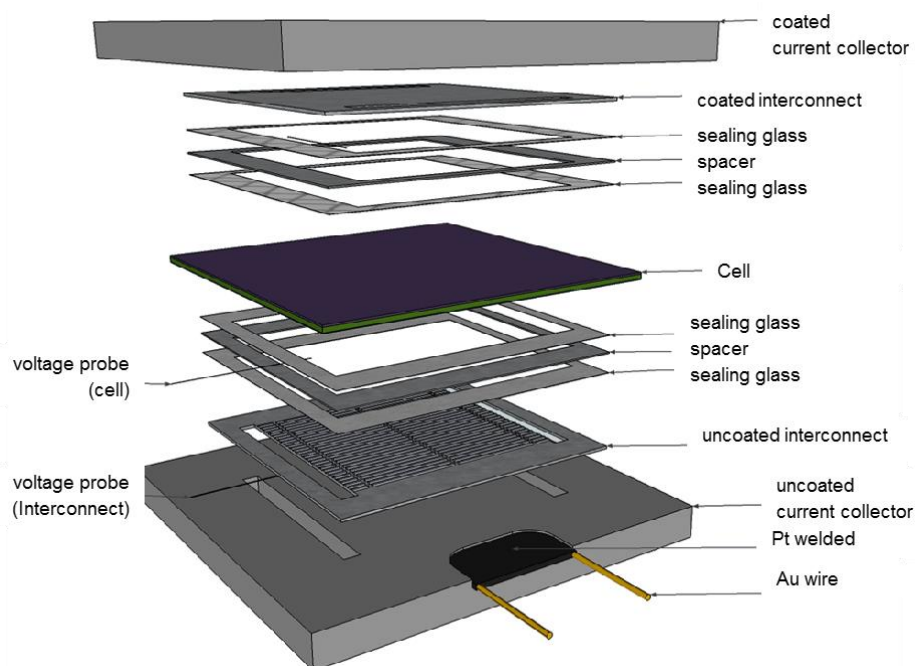


Figure 8. Diagram of the developed stack-element test configuration for accelerated lifetime testing.

One of the challenges in these tests is the insertion of the voltage probes (avoiding gas leakage, bypassing of gases and/or loss of contact while mounting) and to ensure sufficient contact between interconnects and electrodes. Performing impedance measurements over the potential probes directly at the cell allows the determination of the electrolyte resistance, like it is done in single cell tests. Measurements performed over the interconnect voltage probes have an additional ohmic contribution from the interconnects. The combination of the measurements can therefore provide further information on the contact resistance between cell and interconnects. This test configuration was used to accomplish milestones M2.3 and M2.4.

Smaller-scale specialized test configurations were developed and used to obtain detailed measurements on individual cell components. Simplified electrodes were used with advanced electrochemical and spectroscopic techniques to investigate electrode kinetics and degradation phenomena, enabling isolation of the response of the single component which is inaccessible in measurements made on the full, complex device [7-9,25,24]. Some of the simplified model electrodes that were used for detailed electrode kinetics measurements are shown in Figure 9. The differences in electrode kinetics with H₂/H₂O vs CO/CO₂ gases on model vs porous electrode geometries was investigated for Ni-based and doped-ceria electrodes using these configurations [7]. Another model electrode configuration (not shown) was used for a detailed mechanistic investigation of a severe degradation mechanism, carbon deposition, by X-ray photoelectron spectroscopy [25].

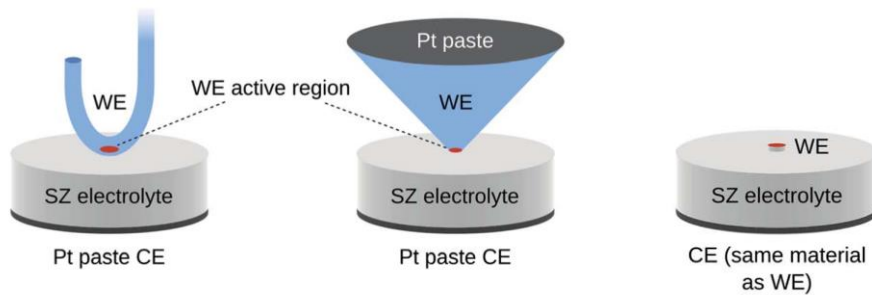


Figure 9. Diagrams of three of the model electrodes employed. WE = working electrode. CE = counter electrode. SZ = stabilized zirconia. From [7].

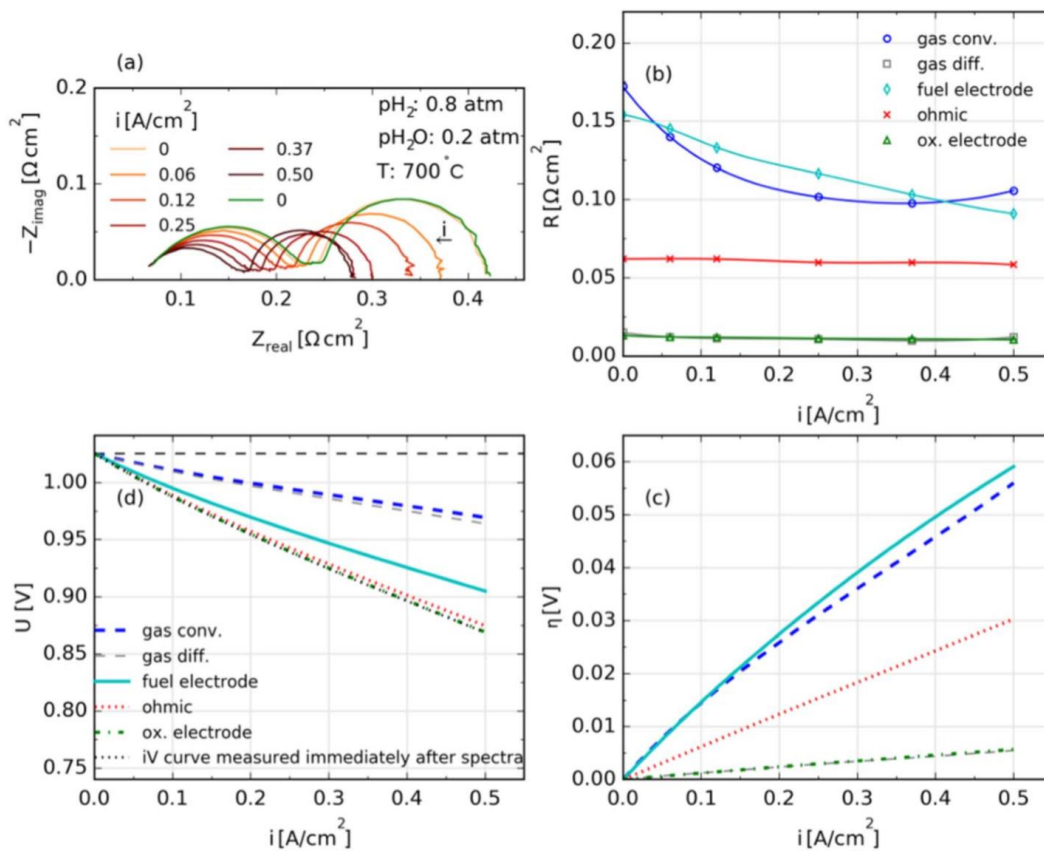


Figure 10. Calculation of overvoltages of individual components from full-cell impedance data. (a) Impedance measurements at different fuel-cell mode current densities. (b) Component-wise resistances from fitting an impedance model. (c) Overvoltages calculated from (b) by integration. (d) Subtraction of overvoltages from open-circuit voltage and comparison with a DC current-voltage curve. From [11].

Detailed impedance spectroscopy measurements on the actual, full cells (Figure 10) were used to quantitatively break-down the cell impedance, via complex nonlinear least squares fitting to a suitable impedance model, into resistances and overvoltages of each individual component (electrodes, electrolyte, gas transport) and link the component measurements to the full-cell geometry [10-12]. The state-of-the-art, fuel-electrode supported planar cells were produced at DTU Energy and were comprised of a 300 μm thick porous Ni/YSZ support on a 12-15 μm thick porous Ni/YSZ electrode and a dense $\sim 5 \mu\text{m}$ thick YSZ electrolyte. A dense $\sim 5 \mu\text{m}$ thick Gd-doped ceria (CGO) barrier layer was deposited by physical vapor deposition, and a lanthanum strontium cobaltite (LSC)-CGO composite oxygen-electrode was applied by screen-printing.

The measurements were then used in 3-D multi-physics models of the full device that were constructed to determine with a new level of accuracy where local “hotspots” of degradation can occur and provide input for improvement of cell design [23,26-27]. Comparison of the multi-physics model with lab measurements showed good agreement (Figure 11) and tuning of modelling parameters can further improve the agreement. A unique aspect of the model not present in many models reported in literature is the inclusion of thermal physics with all basic forms of heat transfer, which has a significant impact on the temperature along the cell. Figure 12 shows the 3-D temperature distribution at two operating points, one in fuel-cell mode in which heat is generated and one in electrolysis mode at an operation point in which heat is consumed. This can be used to determine the possible impact of rapidly changing operating points, which can clearly cause substantial thermal cycles.

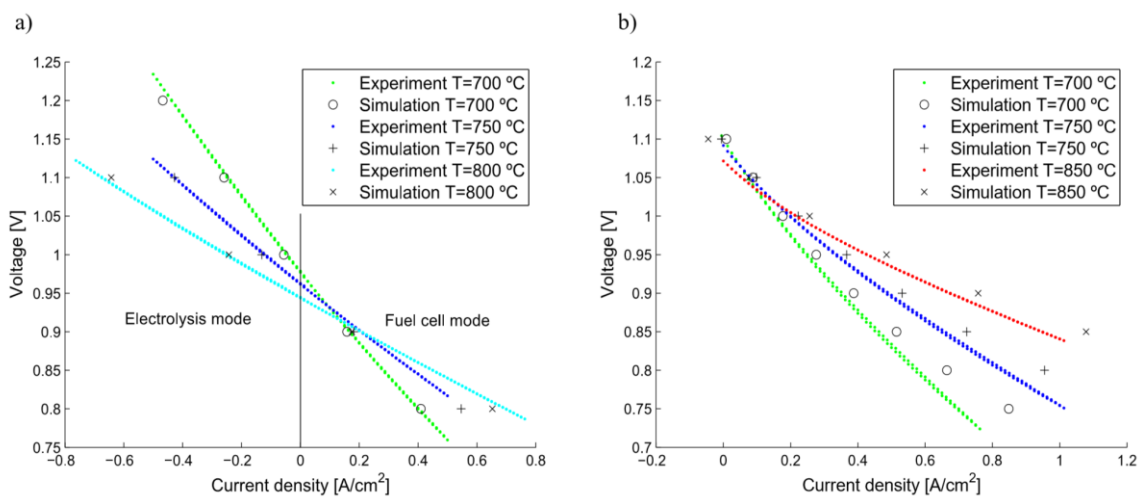


Figure 11. Comparison of simulated and experimentally measured current density vs voltage curves at different temperatures for (a) 50%/50% $\text{H}_2/\text{H}_2\text{O}$ gas supply in electrolysis and fuel-cell modes, (b) 96%/4% $\text{H}_2/\text{H}_2\text{O}$ gas supply in fuel-cell mode. From [26].

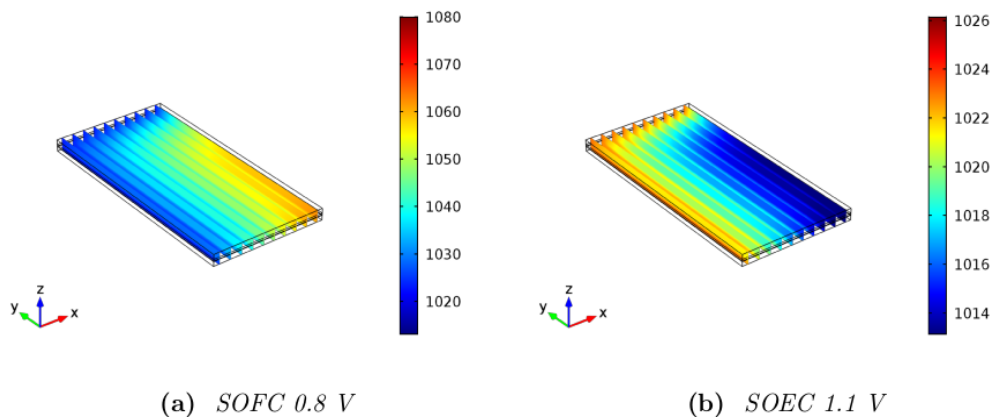


Figure 12. 3-D temperature distribution (in Kelvins) modelled for (a) exothermic fuel-cell mode operation and (b) endothermic electrolysis mode operation.

WT2.2 – Realistic operation

Planar, anode-supported full cells – with layer compositions and thicknesses described earlier in WT2.1 – were operated at constant conditions at 700 °C for more than 2 and 1 years with hydrogen and internally-steam-reformed methane fuel supplies, respectively (Figure 13). These tests were started near the end of the previous project “Towards Smart Grid Ready SOFC” (ForskEL project no. 2012-1-10747) and continued in this project. The results provide baseline data and highlight the need for such “very long term” tests, since the degradation profile changes drastically at about 1000 h of operation, which has typically been the total test length reported in literature and in previous projects. These tests used clean nickel and gold contact components on the fuel and air sides, respectively, and provided baseline stability data with degradation rates as low as 2.5 mV per 1000 h (0.3% voltage drop) during 1 year of operation, accelerating degradation during the test, and faster degradation for the methane test. In both cases, impedance spectroscopy showed that the Ni-YSZ electrode was the main source of performance loss.

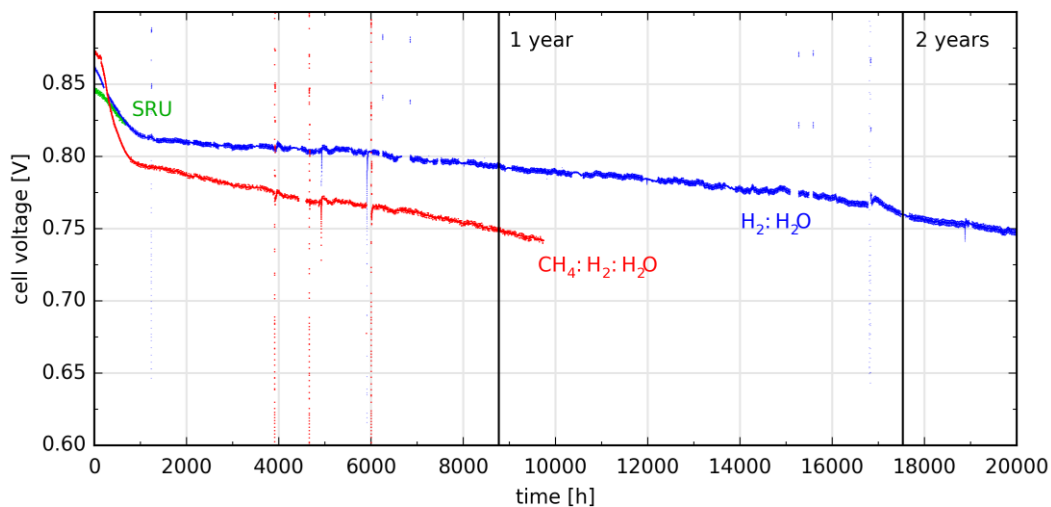


Figure 13. Very long-term baseline cell tests operated at 700 °C at 0.5 A/cm² and SRU test at 750 °C at 0.5 A/cm². The H₂:H₂O test was with 80%/20% H₂/H₂O fuel gas supply and air as oxidant. The CH₄ test was with 30%/10%/60% CH₄/H₂/H₂O fuel gas supply and air as oxidant. The SRU test was with 75%/25% H₂/H₂O fuel gas supply and O₂ as oxidant.

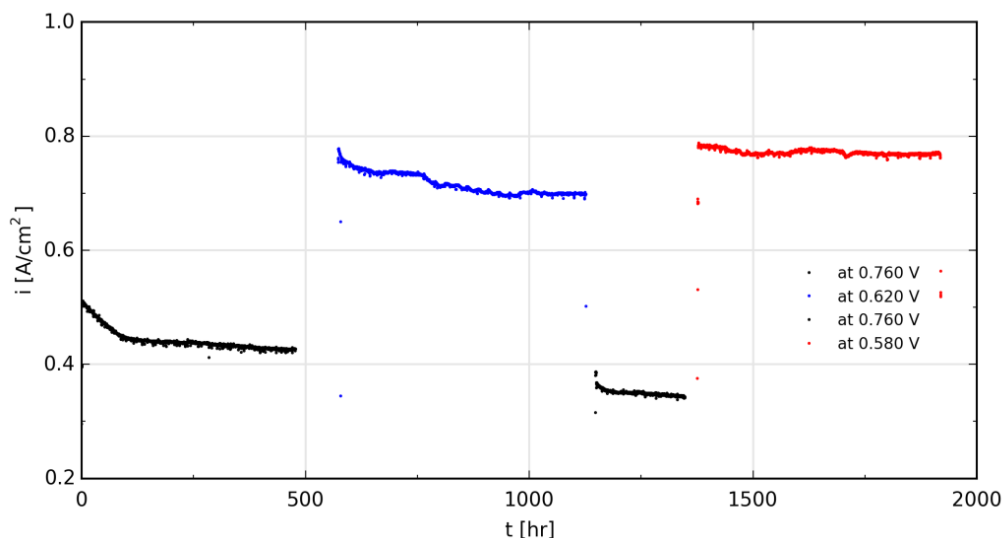


Figure 14. Current density measured at different cell voltages in a constant-voltage mode test.

Stack-element testing using steel contact components was also carried out, both for comparison as well as for accelerated testing. The slope of the cell voltage during the first 1000 h of a SRU test is remarkably similar to that of the cell tests (Figure 13), indicating that the steel components did not cause any increased degradation during this period, although the test conditions were slightly different and the oxygen-electrode was lanthanum strontium manganite based instead of LSC-based.

Another type of cell test that is relevant for realistic operating scenarios is constant-voltage operation. Figure 14 shows the current density during a baseline constant-voltage cell test and illustrates that the degradation profile changes to a slower rate after a similar time period to the constant-current tests – in the range of 1000 h. Interestingly, the cell does not appear to degrade more quickly at lower cell voltages (which correspond to larger overvoltages).

WT2.3 – Accelerated testing

Long-term stack-element testing was carried out to investigate steam content in the fuel gas as a possible degradation acceleration parameter. For the long-term TEST A and TEST B the following testing conditions were chosen: For cell TEST A, 6 different testing sequences during a continuous durability test at 750°C were performed (for details see Table 1). In between each testing period a short characterization with different fuel/oxidant compositions via EIS at OCV conditions has been conducted. In cell TEST B testing sequence 6 of long-term TEST A was conducted right after the reduction and the initial characterization.

Table 1. Conditions for accelerated tests Test A and B. FU = fuel utilization.

sequence	t / h	j / A cm ⁻²	p(H ₂ O) _{in}	p(H ₂ O) _{out}	v(H ₂ O) out / mol h ⁻¹	FU / %	oxidant
TEST A (1)	1000	0.5	0.25	0.53	0.28	37	O ₂
TEST A (2)	1000	0.5	0.25	0.36	0.49	15	O ₂
TEST A (3)	300	0.5	0.25	0.53	0.28	37	O ₂
TEST A (4)	350	0.5	0.25	0.81	0.22	74	O ₂
TEST A (5)	300	0.5	0.4	0.85	0.28	74	O ₂
TEST A (6)	300	0.5	0.72	0.93	0.69	74	O ₂
TEST B	300	0.5	0.72	0.93	0.69	74	O ₂

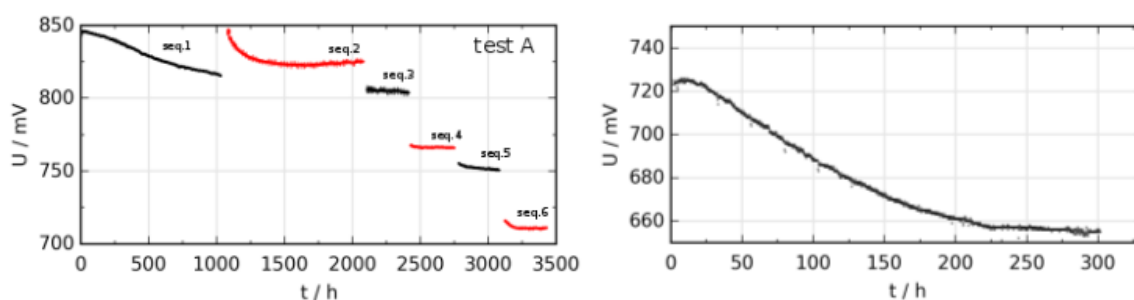


Figure 15. Cell voltage degradation of (left) TEST A at 0.5 Acm⁻² and T = 750°C, flow rate and FU are varied for each sequence (see **Error! Reference source not found.**); (right) TEST B at T=750C, p(H₂O)=0.72, FU=74%.

Based on detailed impedance spectroscopy and scanning electron microscopy (not shown), it was found that steam content in the fuel gas showed a strong effect on degradation of “fresh” Ni-YSZ fuel-electrodes (during the initial testing period) but little effect on an already-aged electrode. Quantitative microscopy data indicates that growth and percolation loss of Ni particles play a key role in the degraded structures.

Two important aspects for accelerated lifetime testing can be abstracted: (i) If using steam as stressor or for accelerated lifetime tests, its threshold and accelerating impact seems to shift with time. While the ex-situ applied testing conditions might have a severe impact on an un-degraded cell of a similar microstructure, this might not be the case for a pre-tested cell. Even a stepwise increase of the apparent stressor does not lead to acceleration in cell TEST A. (ii) Furthermore, by staying below the allowed stressor threshold in the initial degradation period, the same degradation result can be achieved by supplying additional steam.

Based on Ni-YSZ component testing and quantitative microscopy, microstructural evolution is determined by the specific initial microstructure: Apparently, a more porous structure allows more Ni-redistribution and favours enlargement of Ni-particles. Furthermore, a more porous structure can be influenced just by the choice of reduction temperature (1000 °C vs 850 °C in this study) and so this parameter additionally needs to be considered for accelerated lifetime testing. This effect is less dominant for denser microstructures and in those cases can be neglected.

A series of long-term dynamic operation tests were carried out to investigate beneficial or detrimental effects on the degradation rate compared with steady-state reference tests [12]. These tests clearly show both beneficial and detrimental effects (Figure 16). The data plotted in green, “steady-state, no gas cleaning”, shows a steady degradation as the cell voltage drops from 0.84 to 0.82 V during the 500 h of test time shown. For the data plotted in red, “cyclic, no gas cleaning”, the cell is cycled between fuel-cell and electrolysis modes, and the degradation rate is orders of magnitude faster, clearly showing a detrimental effect of this particular set of dynamic operating conditions. Electrolysis mode is more sensitive to impurities in the gas supply and lower temperature operation also increases the sensitivity. The data plotted in blue, “cyclic, gas cleaning”, on the other hand, exhibits zero degradation during the same time 500 h time period, clearly showing a beneficial effect.

Therefore, long-term dynamic operation in reversible mode (periodic switching between fuel-cell and electrolysis modes) revealed that gaseous impurities cause fast degradation, which does not occur in steady-state fuel-cell mode. With a cleaned fuel gas supply, operating in reversible mode showed much lower degradation than in the steady-state fuel-cell mode without gas cleaning [12].

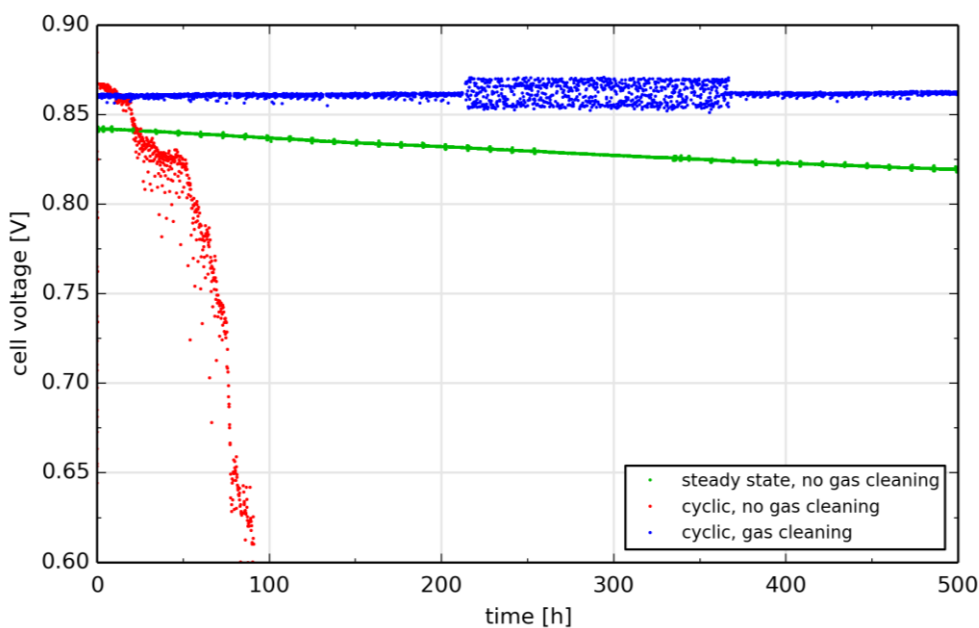


Figure 16. Initial segments of three dynamic operation tests

All 3 tests were carried out at 700 °C, with a gas mixture of either H₂:H₂O 60:40 or 50:50 supplied to the fuel-electrode and air supplied to the oxygen-electrode, and operated at either +0.5 A/cm²

(steady-state) or $\pm 0.5 \text{ A/cm}^2$ (cyclic), where positive current density corresponds to fuel-cell mode and negative corresponds to electrolysis mode. Figure 17 shows the same cyclic tests data as above but including also the higher-voltage electrolysis mode part of the data.

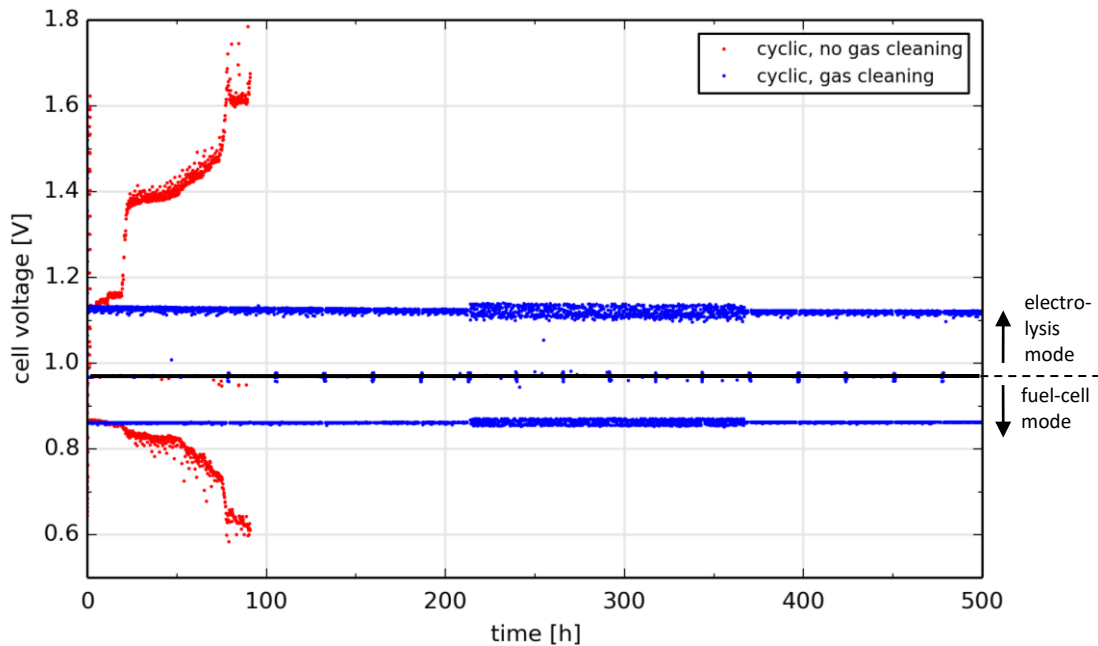


Figure 17. The same initial segments of three dynamic operation tests as in Figure 16, showing also the electrolysis mode part of the data

3.4. WP3 – Improving lifetime and efficiency

WP3 investigated ways to improve lifetime and efficiency by: *in-situ* repair/regeneration during operation, *ex-situ* repair/regeneration, and development of new and improved cells.

WT3.1 – *In-situ* repair/regeneration to extend lifetime

In-situ regeneration is a new concept that entails careful adjustment of operating conditions (gas composition or applied voltage) to regain performance, usually of electrodes [5]. We tested high cathodic polarization of Ni-YSZ electrodes [29] as well as oxidation/reduction cycles of fuel-electrodes [13,28], both of which were able to restore performance by regaining reaction site density.

For the high cathodic polarization study, a series of five tests conducted on state-of-the-art Ni/YSZ-YSZ-CGO_{barrier}-LSC/CGO based single cells. All cells were deliberately degraded via gas stream impurities in CO₂/CO or harsh steam electrolysis operation. Subsequently, the cells were re-activated via a variety of high cathodic polarization or reverse current treatments (RCTs). Electrochemical impedance spectroscopy and scanning electron microscopy showed that the Ni/YSZ electrode performance could be recovered via RCT by producing active nano-particles in the fuel electrode which decrease the charge transfer resistance. It was possible to obtain a lower Ni/YSZ electrode resistance than the initial resistance. The application of the RCT lead to a reduction of the total fuel electrode resistance until an asymptotic trend was attained after which any further RCT pulse was not beneficial. Typically, 5-14 cycles of RCT were required to reach the highest fuel electrode performance (Figure 18). Detrimental effects at the fuel-electrode/electrolyte interface were also observed when applying RCT cycles, leading to an increase in ohmic resistance. Therefore, a tuning of RCT parameters (pulse current density, hold time, number of cycles) is required to avoid detrimental ohmic resistance increase, which offsets the beneficial activation connected to the nano-particles formation.

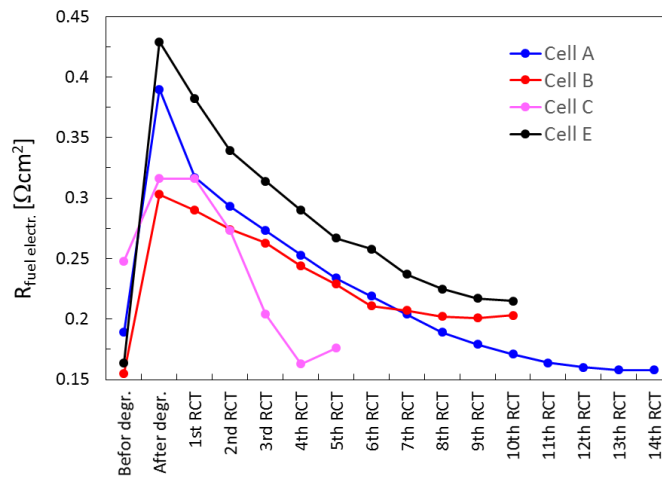


Figure 18. Ni-YSZ electrode resistance evolution through degradation and subsequent re-activation by RCTs, quantified by fitting full-cell impedance data to a suitable model. From [29].

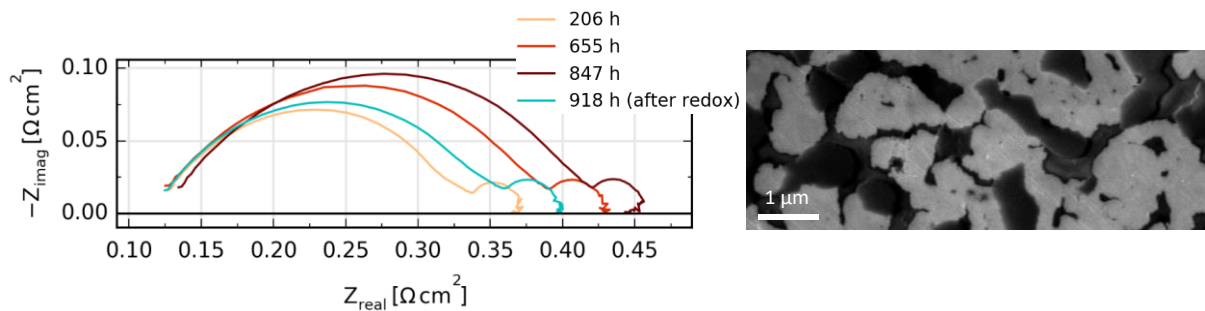


Figure 19. Re-activation of a Ni-YSZ-supported cell by a redox cycle. (left) Impedance spectra measured before and after the redox cycle, at 700 °C, 50%/50% H₂/H₂O vs air. (right) Scanning electron micrograph of a Ni-YSZ electrode structure after a redox cycle.

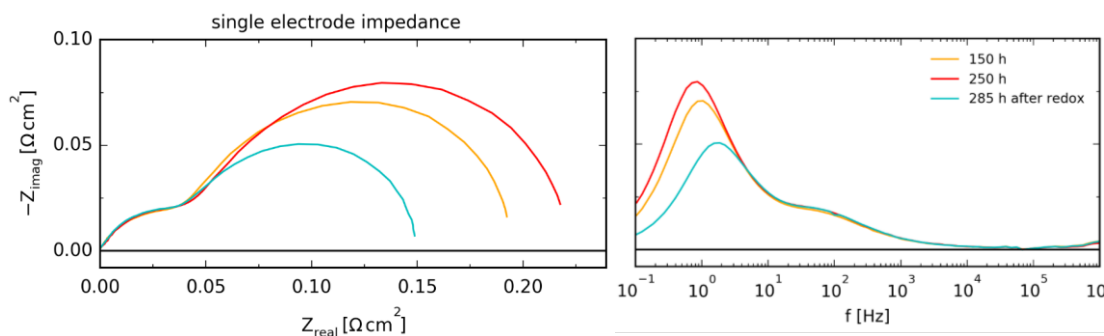


Figure 20. Reactivation of a ceria-based electrode by a redox cycle. Impedance spectra measured at 700 °C, 80%/20% H₂/H₂O vs air.

A second *in situ* method to counteract performance loss by adjustment of operating parameters that was investigated was the brief oxidation of a fuel-electrode by exposure to air followed by re-reduction (a “redox” cycle). For Ni-YSZ electrodes, the large volume expansion during Ni oxidation can be damaging (Figure 3). However, careful control of the redox cycle parameters can lead to activation by refreshing the 3-phase-boundary reaction sites (Figure 19) – somewhat similar to the RCTs.

This appears to be a promising lifetime-extension method but should be investigated in more detail and should be used with caution due to the possibility for causing cell failure.

New nickel-free, ceria-based electrodes that were developed in this project were also re-activated by redox cycles (Figure 20). These electrodes [13,14,28] have internal surfaces that are coated with ceria nanoparticles and are described further in WT3.3. The precise mechanism by which this redox re-activation works is not yet clear. Unlike nickel, CGO does not undergo a phase change, but the chemical expansion and contraction from changes in oxygen stoichiometry cause small volume changes which we believe can create micro/nano-cracks and thereby regain surface area that was lost by coarsening. This redox re-activation was also demonstrated for the first time with full cells (not shown). At both the single-electrode and full-cell level, the redox re-activation could be repeated many times.

WT3.2 – Ex-situ repair/regeneration to extend lifetime

Ex-situ regeneration of Ni-YSZ fuel-electrode performance was done by cooling down a cell, coating the porous fuel-electrode with nanoparticles and heating back up [2,30]. After degradation, cells were cooled, taken out of the test house, and the fuel side was infiltrated with precursor nitrates solution and heat-treated at 350 °C/0.5 h ex-situ in air to form CGO nanoparticles. As seen in Figure 21, reasonable nanoparticle coverage is obtained all the way to the electrolyte. After a cleaning of the electrode surface, the cell was then re-mounted in the test house and heated to operation temperature. The performance improvement is quantified in Figure 22. The fuel-electrode resistance was reduced by 25% in both H₂/H₂O and CO/CO₂ at 750 °C. This promising method was further investigated by a series of systematic tests and at stack level (not reported in this project).

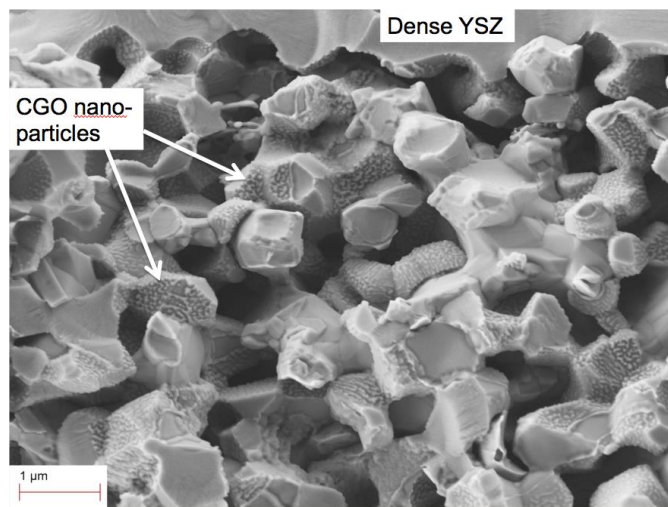


Figure 21. Microstructure of the infiltrated Ni-YSZ fuel electrode

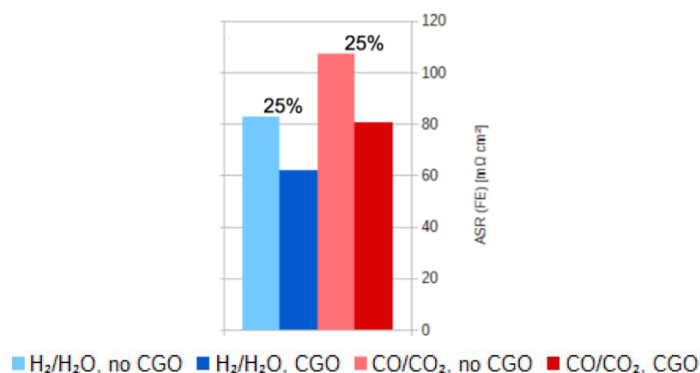


Figure 22. Area-specific resistance (ASR) of the Ni-YSZ fuel-electrode (FE) before and after CGO infiltration.

WT3.3 – Improved cells

Completely new cells were developed that did not contain nickel in the fuel-electrode. By removing nickel, the source of many modes of degradation, a cell with more robust electrodes was obtained. First, new fuel-electrodes comprised mainly of nanostructured Gd-doped CeO₂ were developed [7,13-15,28,31,32]. They provided a polarization resistance as low as 0.01 Ω cm² at 600 °C in hydrogen/steam fuel environment (Figure 23), an order of magnitude lower than conventional nickel-based electrodes [14]. For practical porous electrodes, Gd-doped CeO₂ provides insufficient electronic conductivity so a small fraction of SrFe_{0.75}Mo_{0.25}O₃ (SFM) was added (Figure 24) and these electrodes were found to have almost as low resistance and they were stable against carbon deposition and oxidation [13,28].

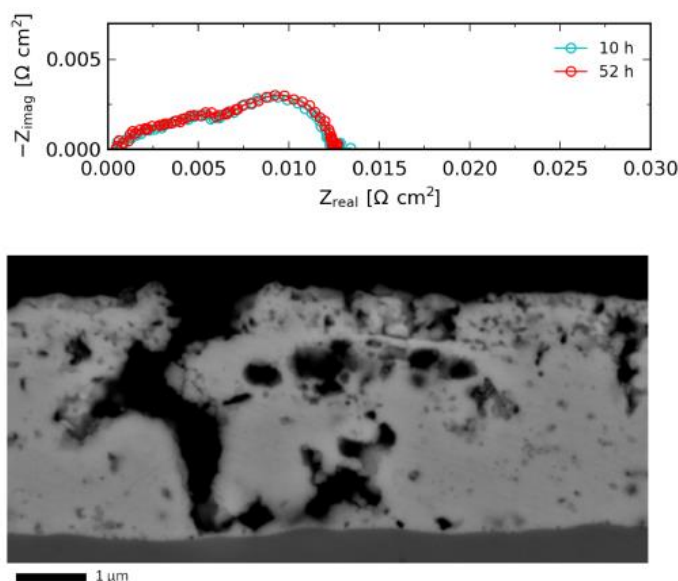


Figure 23. High surface area nanostructured CGO thin-film electrodes. (top) Impedance spectra showing unprecedented low electrode resistance at 650 °C in 80%/20% H₂/H₂O. (bottom) Scanning electron micrograph of thin-film electrode structure.

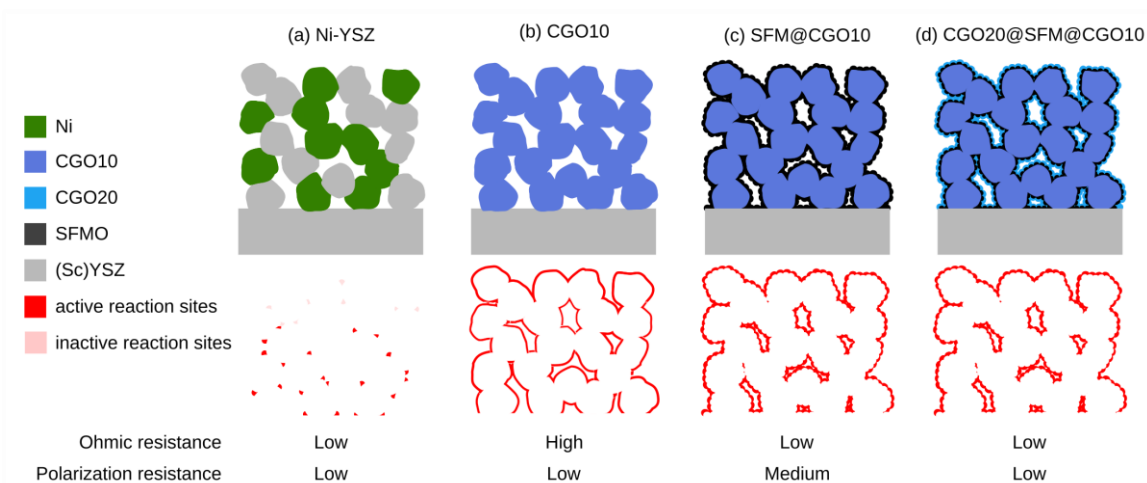


Figure 24. Illustrations of (a) a Ni-YSZ composite, (b) a CGO10 electrode, (c) a CGO10 electrode infiltrated with SFM and (d) a CGO10 electrode infiltrated with SFM and CGO20. Qualitative depiction of reaction sites and estimations of the ohmic and polarization resistance are also shown. CGO10 and CGO20 correspond to CeO₂ with 10% and 20% respectively of Gd doped on Ce lattice sites. From [14,28].

Since the Ni-YSZ is the usual support-layer for thin-electrolyte cells and the goal was to remove nickel, oxygen-electrode-supported cells were developed. Stable and high-performance oxygen-electrodes that do not contain cobalt, with Pr-doped CeO₂ (CPO) as an electrocatalytic material, were also developed. In fact, PDC was used in both fuel and oxygen electrodes [14]. To ensure good structural strength, a YSZ backbone has been employed for the OSC support. The backbone is infiltrated with precursor solutions of Sr-doped LaMnO₃ (LSM) and CPO which yield nanoparticulate coatings on the YSZ backbone that provide electronic conductivity and electrocatalytic activity. LSM is well known to be stable but does not provide as high performance as cobaltite oxygen-electrodes. With the addition of CPO, the electrodes show performance almost as good as the best cobaltite electrodes. Remarkably, we have found that these oxygen-electrodes are completely stable for more than 1000 h (Figure 25).

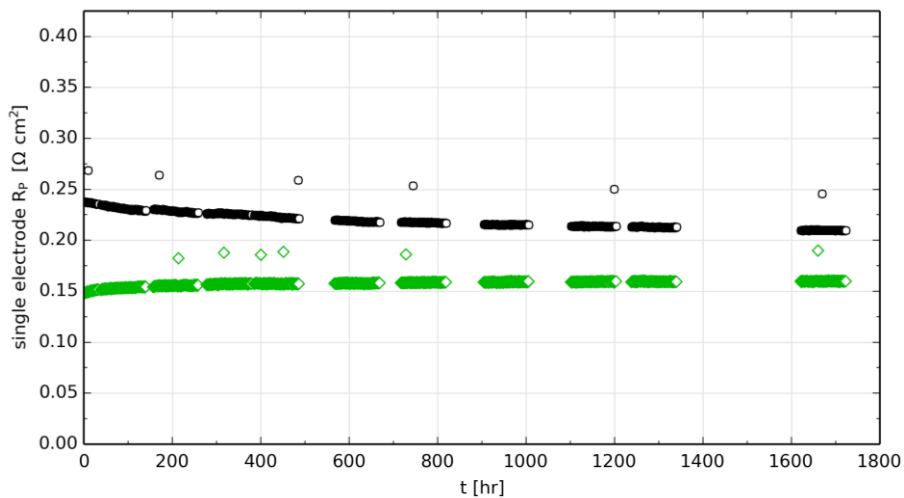


Figure 25. Long-term stability of the polarization resistance of oxygen-electrodes comprised of LSM (black circles) and CPO-LSM (green diamonds) infiltrated YSZ backbones.

Full 5x5 cm² cells made with these fuel and oxygen-electrodes showed equivalent electrode performance and higher stability than state-of-the-art Ni-YSZ supported cells. We found that carbon deposition and oxidation treatments caused no damage, and actually the latter improved the fuel-electrode performance (as shown earlier in WT3.1), suggesting that these new cells could be a game-changer for SOC technology commercialization. For future scale-up purposes, electrode backbone structures made by freeze-casting were also developed [16].

One of these new 5x5 cm² cells was tested long-term in carbon deposition conditions in the project ForskEL 2015-1-12276 "Towards Solid Oxide Electrolysis Plants in 2020" and showed much higher stability than conventional Ni-YSZ-supported cells. This new type of cell is currently being further developed and optimized in subsequent projects.

3.5. Publication and dissemination

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Articles submitted for publication or in preparation (8):

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27. M. Navasa, H.L. Frandsen, T.L. Skafté, B. Sundén, C. Graves, *Localized Carbon Deposition in Solid Oxide Electrolysis Cells Studied by Multiphysics Modeling*, In preparation.
28. T.L. Skafté, B.R. Sudireddy, P. Blennow, C. Graves, *Highly active, carbon deposition resistant and redox stable oxide fuel-electrodes with multi-coated backbone structure for solid oxide cells*, In preparation.
29. A. Hauch, M. Marchese, A. Lanzini, C. Graves, *Re-activation of degraded Ni/YSZ SOFC anodes – Nano-particle formation via reverse current pulses*, Submitted.
30. T.L. Skafté, J. Hjelm, P. Blennow, C. Graves, *Eliminating fuel electrode degradation and re-activating solid oxide cells and stacks*, Submitted.
31. C. Graves, L. Martinez, B.R. Sudireddy, *Exceptional activity of nanostructured ceria films prepared by metal-organic solution deposition for electrochemical H₂O reduction and H₂ oxidation*, In preparation.
32. L. Martinez, B.R. Sudireddy, C. Chatzichristodoulou, C. Graves, *The role of nickel in nanostructured doped-ceria thin-film electrodes for solid oxide cells*, In preparation.

Presentations (11):

33. T.L. Skafté, **Presentation of paper #1 above**, at the 12th European SOFC & SOE Forum (EFCF2016), July 5-8, 2016, Lucerne, Switzerland
34. C. Graves, **Presentation of paper #5 above**, at Royal Society of Chemistry (RSC) Faraday Discussion “Solid Oxide Electrolysis: Fuels and Feedstock from Water and Air”, July 13-15, 2015, York, UK
35. J.-C. Njodzefon, **Presentation of paper #6 above**, at the ECS Conference on Electrochemical Energy Conversion & Storage with Solid Oxide Fuel Cells 14 (SOFC-XIV), July 26-31, 2015, Glasgow, UK
36. J.-C. Njodzefon, **Presentation of paper #8 above**, at 2014 ECS and SMEQ Joint International Meeting, October 5-9, 2014, Cancun, Mexico
37. A. Ploner, **Presentation of paper #11 above**, at the 12th European SOFC & SOE Forum (EFCF2016), July 5-8, 2016, Lucerne, Switzerland
38. T.L. Skafté, **Presentation of paper #13 above**, at the 229th Meeting of the Electrochemical Society, May 29-June 2, 2016, San Diego, California
39. C. Graves, **Invited presentation of paper #14 above**, at the 229th Meeting of the Electrochemical Society, May 29-June 2, 2016, San Diego, California
40. M. Mogensen, **Presentation of paper #17 above**, at the 229th Meeting of the Electrochemical Society, May 29-June 2, 2016, San Diego, California
41. M. Navasa, **Presentation of paper #27 above**, at the 230th Meeting of the Electrochemical Society / PRiME 2016, October 2-7, 2016, Honolulu, Hawaii
42. J. Nielsen, **Presentation of paper #23 above**, at the 230th Meeting of the Electrochemical Society / PRiME 2016, October 2-7, 2016, Honolulu, Hawaii

43. V. Gil, *On the challenges of developing high-performance porous structures*, at Energy, Materials, and Nanotechnology (EMN) Meeting on Ceramics 2016, January 28, 2016, Hong Kong, China

3.6. Environmental benefits of the project

Denmark is committed to contribute to mitigate global climate change per the United Nations Framework Convention on Climate Change. An integral part of this commitment is the reduction of greenhouse gas emissions in Europe by 80-95% in 2050 compared to the 1990 level. Reversible solid oxide fuel cells are a potential key technology to achieve CO₂ emissions reductions by facilitating integration of a higher fraction of intermittent renewable energy in the electricity grid.

SOFCS can provide dispatchable power generation to fill in the gaps of wind/solar supply. Since they can be operated reversibly, in electrolysis mode, they can also provide electrical energy storage like a battery. Compared with conventional batteries, storing energy in the bonds of carbon and hydrogen results in far lower usage of toxic metals (e.g. Co, Ni) per kWh stored. Another way the same technology can enable a fossil free future is by efficiently electrolyzing H₂O and CO₂ to ultimately produce synthetic hydrocarbon fuels that can directly displace fossil hydrocarbons (such as natural gas and oil).

This project contributes directly to these environmental benefits by making significant progress towards achieving a commercial breakthrough with the solid oxide cell technology via considerable improvements in performance and durability.

4. Utilization of project results

The results achieved in the project will be utilized in DTU Energy's various industrial and academic partnerships. DTU Energy collaborates with a number of SOFC and SOEC companies and research institutes that share goals in achieving improved performance and durability of the technology – targets this project has achieved in new ways. Haldor Topsoe A/S (HTAS), for example, is commercializing a CO₂ electrolyzer using SOECs and using the new cells developed in this project, which are stable against carbon deposition, is expected to be of high interest for HTAS's electrolyzer product. In fact, DTU Energy and HTAS have already submitted joint proposals on this topic. Other European industrial partners like SolidPower and Sunfire, with whom DTU Energy has ongoing EU projects, are expected to benefit from communication of these results. Furthermore, the results will be the basis for further research and development at DTU Energy Conversion.

5. Conclusion and outlook

The key outcomes of the project can be summarized as follows:

1. A comprehensive review of lifetime-limiting factors that provides a quantitative analysis of the current international status of degradation and lifetime in the field [1-4] and identification of the most important factors and re-activation mechanisms [5].
2. A techno-economic analysis of a new market scenario utilizing a single device in reversible operation mode – buying electricity and selling methane fuel product, or vice versa, when the market price spread was sufficient to obtain revenue – using realistic performance and durability data, system design, operating profiles, and Denmark grid market data, which showed that reversible operation is promising for future profitability [6].
3. New understanding of electrode kinetics and degradation phenomena using simplified electrodes with advanced electrochemical and spectroscopic techniques [7-9,25,24].

4. 3D multi-physics models of cells that were used to determine with a new level of accuracy where local “hotspots” of degradation can occur and provide input for improvement of cell design [23,26-27].
5. Very long-term single-cell tests operated for more than 2 years showed the long-term stability profile and confirmed the importance of testing beyond 1000-2000 h.
6. Development of a new stack-element testing platform which did not introduce any additional degradation compared with the idealized single-cell tests.
7. Steam content in the fuel gas was used as an acceleration vector and showed a strong effect on degradation of “fresh” Ni-YSZ fuel-electrodes (during the initial testing period) but little effect on an already-aged electrode.
8. Long-term dynamic operation in reversible mode (periodic switching between fuel-cell and electrolysis modes) revealed that gaseous impurities cause fast degradation, which does not occur in steady-state fuel-cell mode. Operating in reversible mode with a cleaned fuel gas supply showed much lower degradation than in the steady-state fuel-cell mode without gas cleaning [12].
9. New *in situ* performance regeneration treatments by high cathodic polarization of Ni-YSZ electrodes [29] or oxidation/reduction cycles of fuel-electrodes [13,28] were both able to restore performance by regaining reaction site density.
10. Demonstration of *ex-situ* regeneration at cell level and for Ni-YSZ electrode performance recovery by cooling down the device, infiltrating nanoparticles in the fuel-electrode and heating back up [2,30].
11. New cells were developed that did not contain nickel in the fuel-electrode which were more robust than state-of-the-art Ni-YSZ based cells, showing complete stability against carbon deposition and oxidation. The new fuel-electrodes based on nanostructured ceria as the active electrocatalytic material also showed higher reaction rates than Ni-YSZ, especially at low temperatures in the 500-700 °C range [7,13-15,28,31,32]. The new electrodes were successfully integrated into full 5x5 cm² cells and tested through carbon deposition and oxidation with much lower performance degradation – in fact activation in some cases – than Ni-YSZ based cells.

The project outcomes strongly suggest that further research especially in the above items #2, 9, 10 and 11 will be highly valuable for progressing the solid oxide cell technology. Besides additional scientific research on these topics, making the new cells (#11) scalable will be an important next step towards a commercially viable, next generation of SOCs. Technology transfer of #10 has already been initiated with one of DTU Energy’s industrial partners, HTAS, via another ForskEL project, and HTAS has also expressed strong interest in the new cells (#11). Overall, the outcome of this project provides guidelines for the future focus of development and research in order to achieve increased performance, improved diagnostics, better durability and robustness, and more cost-efficient solid oxide cell technology, which promises to be a key technology for enabling a high fraction of intermittent wind and solar power supplies.