Test and characterization of reversible solid oxide cells and stacks for innovative renewable energy storage

Hauch, Anne; Ploner, Alexandra; Pylpko, Sergii; Mougin, Julie; Cubizolles, Géraud

Published in:
Proceedings of the 14th European SOFC & SOE Forum 2020

Publication date:
2020

Document Version
Publisher's PDF, also known as Version of record

Citation (APA):
Test and characterization of reversible solid oxide cells and stacks for innovative renewable energy storage

Anne Hauch* (1), Alexandra Ploner (2), Sergii Pylpko (3), Julie Mougin (4), Geraud Cubizolles (4)
(1) Dept. of Energy Conversion and Storage, Technical University of Denmark
2800 Kgs. Lyngby/Denmark
(2) SGL Carbon GmbH, 86405 Meitingen/Germany
(3) Elcogen AS, 11415 Tallinn/Estonia
(4) Univ. Grenoble Alpes, CEA/LITEN
17 rue des Martyrs, 38054 Grenoble Cedex 9/France
*) Tel.: +45-21-36-28-36
hauc@dtu.dk

Abstract

This work aims at developing an innovative renewable energy storage solution, based on reversible Solid Oxide Cell (rSOC) technology. That is to say, one system optimized to operate either in electrolysis mode (SOEC) to store excess electricity to produce H₂, or in fuel cell mode (SOFC) when energy needs exceed local production, to produce electricity and heat again from H₂ or any other fuel locally available. Firstly, work focused on optimization of the different layers constituting the single SOC cell to reach high initial performance applying state-of-the-art materials as previously reported [1]. Secondly, the initially highest performing cells were selected for long-term reversible SOFC/SOEC single cell tests. Thirdly, these cells were integrated in a stack design optimized for reversible operation at high degrees of H₂ and H₂O utilization. The long-term single cell tests showed significant degradation in galvanostatic test periods during electrolysis but not in fuel cell mode prior to starting the reversible test operation while the degradation diminished during the subsequent rSOC operation of the cells operating at 700°C, +0.6 and -1.2 A/cm² in SOFC and SOEC modes respectively, at fuel utilization (FU) up to 80% in both modes. Electrochemical impedance spectroscopy analyses and post-mortem SEM investigations of tested single cells reveal that the fuel electrodes degraded significantly during the long-term single cell tests. Furthermore, long-term stack tests were conducted on 5-cell stacks, integrating both reference cells and optimised cells. The long-term stack tests were conducted applying different switches between SOFC and SOEC modes. Initially long duration tests (100h each mode) were performed in a mixture of 50% H₂O and 50% H₂ to see the effect of the polarisation only. The alternating cycle SOEC/SOFC was repeated over a 1800 h testing period. Then stack switched daily from SOEC mode (8h in 90% H₂O and 10% H₂ @ 0.84 A/cm² FU 54%) to SOFC (16 h in 100% H₂ @ 0.26A/cm²@ FU 60%) for full cycles over a testing period of 500 h. Stack tests were operated in reversible mode for up to several thousands of hours and results on the stack performance and lifetime will be presented.
Introduction

With Europe’s increasing share of electrical energy supply from renewable – but also fluctuating – energy sources such as wind and solar, the need for efficient and flexible energy conversion and storage increases in order to be able to incorporate the renewable energy in the energy grid and ensure security of supply. In this context reversible solid oxide cells (rSOC) can provide flexible and up-scalable solutions for conversion and storage of renewable energy; and this paper will focus on flexible load cycling operation of both single cells and stacks.

1. Scientific Approach

Besides degradation aspects in SOFC and SOEC mode, respectively; reversible operation of SOC additionally involves inversion of electrical fields (i.e. potential gradients), rapid shifts in gas composition and alternation between exo-, thermoneutral and endothermic operation. Cells and stacks need to be designed, optimized and manufactured to cope with these rapidly changing conditions to ensure long-term stability and robustness of the rSOC. Furthermore, high system efficiency requires high (>80 %) reactant conversion rates. With these objectives in mind, this study aims to investigate durability at load cycling operation and optimized stack design for such reversible operation.

The reported work and results are part of the European project REFLEX (www.reflex-energy.eu). Reversible solid oxide cells (rSOC) has been manufactured, optimized, tested and characterized as single cells. Stack design has been optimized for these cells and stacks have been tested in SOFC/SOEC modes in terms of performance and with several types of cycles. This work reports the joined efforts between Elcogen (cell manufacturing), Technical University of Denmark (cell testing and characterization) and CEA (stack design optimization, manufacturing and testing). Stacks will later be incorporated by Sylfen as part of their Smart Energy Hub which also includes a battery system.

2. Experiments

Cell characteristics:
The reference structure (so-called 400-B-SM) for an Elcogen cell is a fuel electrode supported cell, consisting of ~400 µm Ni/3YSZ support layer having a ~12 µm Ni/8YSZ active layer, a ~7 µm thick LSC oxygen electrode, a ~2µm 8YSZ electrolyte and a CGO barrier layer of similar thickness. Different cell microstructures have been produced by Elcogen for this project. The single cells are cut in dimension of 53x53 mm\(^2\) having an active oxygen electrode area of 40x40 mm\(^2\). The initial performance via electrochemical impedance spectroscopy (EIS) of these cells were previously reported and compared [1]. Based on this study two optimized cells were selected for further durability testing, namely the cells denoted “ALD” and “RND14” in [1]. The ALD cell has a thin (sub-micrometer scale) atomic layer deposition (ALD) barrier layer. Both cells have a thicker fuel electrode and oxygen electrode layer compared to reference cells produced by Elcogen.

Single cell test and characterization:
The testing procedure started by heating the single cells at 850°C for sealing and reduction. The NiO of the support and fuel electrode was reduced for 2 h in a 5:95 H\(_2\):N\(_2\) gas mixture at 20 l/h and completed for 1 h in pure H\(_2\) at 20 l/h. After reduction, the cells were initially characterized by iV-curves and EIS at open-circuit voltage (OCV) at 750°C, 700°C, 650°C. For EIS a Solartron 1252A frequency analyzer was used for performance comparison and impedance monitoring over time. The spectra were recorded in the frequency range from 96.5 kHz to 0.1 Hz with 12 points per decade. An in-house developed, Python based software (‘Ravddaw’) was used to analyse and visualize measured EIS spectra [2]. Distribution of relaxation times (DRT) was used for qualitative interpretation of characteristic frequency ranges for different processes [3]. Complex-non-
linear-square (CNLS) fitting to an equivalent circuit was applied for quantification of the impedance of the individual physical/electrochemical processes. For pristine cells, the frequency range between 11-40 kHz can be assigned to the ionic transport/rail in the YSZ-matrix of Ni-YSZ electrodes [4] whereas electrochemical charge-transfer processes of the fuel electrode impedance appear at 1-4 kHz and impedance response in the frequency range 0.1-0.3 kHz can be expected for the applied type of oxygen electrode, respectively. Impedance originating from gas diffusion and gas conversion appear in a frequency range below 100 Hz [4,5].

Galvanostatic durability tests were operated over hundreds of hours. The tests were operated at 700°C and 0.6 A/cm² supplying dry H₂ in SOFC mode for 250 h and subsequently at -1.2 A/cm² at H₂O/H₂:90/10 H₂O/H₂ in SOEC mode for 250 h. The fuel utilization (FU) was 80% in both operating modes and synthetic air was fed to the oxygen electrode. Hereafter load cycling test was started. The reversible load cycling testing was conducted by cycles of 16 h operation in SOFC mode followed by 8 h operation in SOEC mode for both cells.

Stack design and test:
The stack considered is made of 25 cells of 120x120 mm², with a 100 cm² active area. Stack design is presented elsewhere [9]. For integration into stacks, RND14 cells (so-called G2) were selected, ALD cells were not for industrial reasons despite their good performances. First a 5-short cell stack has been manufactured by CEA, and its performance was compared to the same stack comprising reference Elcogen 400-B-SM cells. iV-curves were recorded in SOEC mode, with 90%H₂O/10%H₂ on fuel side, air on the other side, at 800, 750 and 700°C. The nominal flow rate was 12 NmL/min/cm². The maximum current recorded during the iV curve was considered when the less performing cell of the stack reached 1.4V.

Then it was tested in durability, and compared to the stack made of reference cells. Stack durability tests were performed at 700°C. A first period of about 800 h of operation was conducted alternating from SOFC to SOEC mode by steps of ~100 h. During this first part, stack was supplied by 50/50 H₂O/H₂ at total flow rate of 12 NmL/min/cm² on fuel side and air (clean and dry) on oxygen side. Tests were operated galvanostatic at 0.35 A/cm², in SOFC mode and -0.58 A/cm² in SOEC.

Then a second stage of operation was performed alternating from SOEC to SOFC by daily cycles of respectively 8h and 16h. This short period rSOC cycling test was operated over more than 250 h. Results of the short period rSOC cycling will not be presented in the current paper.

Finally, a 25-cell stack made of G2 cells was manufactured. In addition to the design optimizations described in [1] and to the integration of G2 cells, this stack integrates a manifold and an electrical insulation for assembly of 4 stacks into a module produced by Sylfen. Its performance was compared to the 5-cell short stack.

3. Results and Discussion

Initial single cell performance (iV-curves):
Figure 1 shows the initial performance via iV-curves for ALD and RND14 at 700°C applying dry H₂ at 6 l/h for the FC iV-curve and a gas mixture of H₂O/H₂:90/10 to the fuel electrode with a steam inlet flow of 9.4 l/h for the SOEC iV-curve. Air was purged to the oxygen electrode. Only initial performance at OCV was reported in [1]. The area specific resistances (ASR) of the cells are given in Table 1. The ALD cell has the highest performance. Notice that for the iV-curves in fuel cell mode sign of fuel starvation is observed at a current density of approximately 0.7 A/cm² corresponding to a FU of 83%. The original target operation within the REFLEX project was a FU of 85%. Considering the
observed start of starvation for the iV-curves in Figure 1; a FU of 85% is expected to be critical for stack operation and will therefore be adjusted for subsequent stack tests. In SOEC mode steam conversion of ~95% was reached. Furthermore; the project target was operation at 0.6 A/cm² and -1.2 A/cm². At these current densities the expected initial cell voltages for the ALD cell will be 883 mV and 1252 mV, and 857 mV and 1301 mV respectively for the RND14 cells. This is in line with desired operation in the vicinity of thermal neutral potential during electrolysis. Satisfying OCV of 884-885 mV was measured at 90% H₂O in H₂; only 2 mV from the theoretical Nernst potential at 692 °C at which temperature the iV-curves were recorded. This indicate a proper sealing of the cell test set-up and minimal gas leakage through the electrolyte even though the electrolyte is only app. 2 µm thick.

Figure 1: Initial iV-curves for ALD (purple) and RND14 (green) cells at 700°C applying a gas mixture of H₂O/H₂:90/10 to the fuel electrode with a steam inlet flow of 9.4 l/h (9.8 Nml/min/cm²) for the SOEC iV-curves and dry H₂ (6 l/h) for the SOFC iV-curves and air to the oxygen electrode.

Table 1: Data from initial iV-curves recorded at 700°C applying a gas mixture of H₂O/H₂:90/10 with a steam inlet flow of 9.4 l/h and dry H₂ (6 l/h) for SOEC and SOFC iV-curves, respectively (air to the oxygen electrode). ASR values obtained as slope of the iV-curve in the region from ± 0.25 A/cm² to ± 0.50 A/cm².

<table>
<thead>
<tr>
<th>Cell</th>
<th>ASR_SOFC [Ωcm²]</th>
<th>ASR_SOEC [Ωcm²]</th>
<th>i at 1.3 V [A/cm²]</th>
</tr>
</thead>
<tbody>
<tr>
<td>ALD</td>
<td>0.256</td>
<td>0.280</td>
<td>-1.27</td>
</tr>
<tr>
<td>RND14</td>
<td>0.300</td>
<td>0.324</td>
<td>-1.20</td>
</tr>
</tbody>
</table>

Single cell durability test – SOFC, SOEC and load cycling:
Figure 2 shows the cell voltage curves over time for the durability test for the ALD and RND14 cell. The durability tests were started by 250 h of constant galvanostatic SOFC test followed by 250 h SOEC test. Hereafter load cycling testing was conducted for 500 h and 1000 h, respectively for the RND14 and ALD cell. Shortening of load cycling test for RND14 cell was due to labs moving, not due to cell failure. For comparison, a durability test without the operation at constant SOFC and SOEC conditions at start of test is given in Figure 2 (previously reported in [1]). From Figure 2 it is evident that operating in constant SOFC mode only leads to limited degradation (27 mV/kh and 69 mV/kh for ALD and RND14 respectively). In contrast; operation in constant SOEC mode for 250 h lead to significant degradation; 790 mV/kh and 937 mV/kh, for ALD and RND14, respectively. Notice, that the fuel electrode overpotential is significantly different for the cells during
SOFC and SOEC mode of operation. CNLS analysis of the EIS recorded at start and end of the two times constant operations was performed applying the equivalent circuit as described in “Experimental” and key numbers are given in Table 2. Here it is observed that the fuel electrode overpotential ($\eta_{\text{fuel}}$) even at start of electrolysis testing is approximately a factor of 5 higher than at start of SOFC testing for both cells.

Table 2: CNLS analysis of the EIS recorded at start and end of the constant SOFC and SOEC operation. Equivalent circuit model as described in “Experimental”. $R_{\text{fuel}}$ includes the two resistance contributions in the frequency range from app. 40 kHz to app. 1 kHz and the fuel electrode overpotential, $\eta_{\text{fuel}}$, is calculated from $R_{\text{fuel}}$.

<table>
<thead>
<tr>
<th>Conditions</th>
<th>$R_s$ ($\Omega$cm$^2$)</th>
<th>$R_{\text{fuel}}$ ($\Omega$cm$^2$)</th>
<th>$R_{\text{ox.+diff.}}$ ($\Omega$cm$^2$)</th>
<th>$R_{\text{conv.}}$ ($\Omega$cm$^2$)</th>
<th>$R_{\text{total}}$ ($\Omega$cm$^2$)</th>
<th>$\eta_{\text{fuel}}$ (mV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>ALD</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SOFC$_{\text{start}}$</td>
<td>0.056</td>
<td>0.053</td>
<td>0.016</td>
<td>0.338</td>
<td>0.463</td>
<td>32</td>
</tr>
<tr>
<td>SOFC$_{\text{end}}$</td>
<td>0.058</td>
<td>0.051</td>
<td>0.028</td>
<td>0.305</td>
<td>0.442</td>
<td>31</td>
</tr>
<tr>
<td>SOEC$_{\text{start}}$</td>
<td>0.063</td>
<td>0.128</td>
<td>0.050</td>
<td>0.236</td>
<td>0.447</td>
<td>154</td>
</tr>
<tr>
<td>SOEC$_{\text{end}}$</td>
<td>0.070</td>
<td>0.280</td>
<td>0.107</td>
<td>0.586</td>
<td>1.043</td>
<td>336</td>
</tr>
<tr>
<td>RND14</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SOFC$_{\text{start}}$</td>
<td>0.085</td>
<td>0.076</td>
<td>0.010</td>
<td>0.310</td>
<td>0.481</td>
<td>46</td>
</tr>
<tr>
<td>SOFC$_{\text{end}}$</td>
<td>0.088</td>
<td>0.089</td>
<td>0.009</td>
<td>0.356</td>
<td>0.542</td>
<td>53</td>
</tr>
<tr>
<td>SOEC$_{\text{start}}$</td>
<td>0.095</td>
<td>0.213</td>
<td>0.066</td>
<td>0.399</td>
<td>0.773</td>
<td>256</td>
</tr>
<tr>
<td>SOEC$_{\text{end}}$</td>
<td>0.152</td>
<td>0.577</td>
<td>0.090</td>
<td>0.891</td>
<td>1.710</td>
<td>692</td>
</tr>
</tbody>
</table>

For previous SOEC test it has been reported that especially the fuel electrode overpotential (or perhaps more precisely the local $pO_2$ in the active fuel electrode) plays an important role in the irreversible fuel electrode degradation, which can also affect $R_s$, during steam electrolysis [6, 7, 8]. Even identical externally set test conditions do not necessarily lead to identical degradation behavior when sister cells are tested in SOFC versus SOEC mode [8].

Figure 2: Durability single cell tests. For comparison a cell test, “REF cell”, in load cycling mode from start of test is shown, previously reported in [1]. Test operated at 700°C, 80% FU, 0.6 A/cm$^2$ and -1.2 A/cm$^2$, respectively in SOFC and SOEC mode.

To illustrate the overall changes upon the long-term testing depicted in Figure 2; the EIS recorded at OCV, 700°C, air and H$_2$O/H$_2$:50/50 before and after durability and load cycling,
is given in Figure 3 for the ALD cell. The ohmic resistance increases significantly (app. 31 mΩcm$^2$) while the polarization resistance increases in an even more severe way, approximately 164 mΩcm$^2$, having the largest change of impedance in the frequency range 1-10 kHz (fuel electrode contribution). OCV of 974 mV (at 695 ºC) and 971 mV (at 701 ºC) was measured at H$_2$O/H$_2$:50/50 and air upon start and end of the durability test in Figure 3 for the ALD cell; which is within 2 mV of the theoretical Nernst potential. This suggests that the 2 µm thin electrolyte has stayed intact. Similar was observed for the RND14 cell.

![Nyquist and DRT plot of EIS raw data recorded before and after the durability test for ALD cell depicted in Figure 2. EIS recorded at OCV, 700°C, air to the oxygen electrode and 50/50 mixture of H$_2$O and H$_2$ to the fuel electrode.](image)

**Figure 3:** Nyquist and DRT plot of EIS raw data recorded before and after the durability test for ALD cell depicted in Figure 2. EIS recorded at OCV, 700°C, air to the oxygen electrode and 50/50 mixture of H$_2$O and H$_2$ to the fuel electrode.

**Initial stack performance (iV curves):**
Figure 4 presents the performance of the two 5-cell stacks comprising either reference cells or G2 cells. In figure 4a it can be seen that at 700°C, the performance of the stack comprising the G2 cells is slightly better than for the stack made of reference cells. Indeed, the maximum current density was -0.95 A/cm$^2$ as compared to -0.88 A/cm$^2$ for the stack with reference cells, the curves being stopped when the worse cell reaches 1.4V. Also the scattering in voltage seems lower for the stack comprising G2 cells. Figure 4b presents the iV curves at 800, 750 and 700°C for the two stacks. For a sake of clarity of the picture, only the average voltage is presented for each stack. At 750°C, like at 700°C and even a bit more, G2 cells lead to a slightly higher performance of the stack, while at 800°C this is not the case. In all cases, these results validates the integration of G2 cells in the stack. G2 cells performances are lower in stack as compared to single test results reported in Figure 1 for similar testing conditions (T and flow rate). As for single cell tests, high steam conversion can be reached with a value close to 90% at 800°C for the flow rate considered.
Figure 4: Initial iV curves in SOEC mode for 5-cell stack comprising reference cells (blue symbols) and for 5-cell stack comprising G2 cells (yellow symbols); 90%H₂O/10%H₂ with a flow rate of 12 Nml/min/cm² provided to the fuel electrode, air on the other side; steam conversion (SC) is provided on secondary x-axis; a) plot of the 5 cells of each stack at 700°C; b) average cell voltage for each stack at 800, 750 and 700°C

Figure 5 superimposes the performance of the G2 5-cell short stack and of the G2 25-cell stack. In figure 5a, all individual voltages for the two stacks are shown at 700°C. It can be noticed that the voltages are nicely superimposed, with, as expected, a scattering of voltages a bit higher for the 25-cell stacks. Due to the recording methodology stopping the iV curve when one cell reaches 1.4 V, the maximum current density of the 25-cell stack is a bit lower than for the 5-cell stack, -0.93 and -0.95 A/cm², respectively. However when looking at the average cell voltage (figure 5b), the difference is insignificant and it can be concluded that the performances of the two stacks are similar. This trend is confirmed at 750 and 800°C, with a very good superimposition of the curve in the linear part of the plot. Only at high current densities and high steam conversion for 750 and 800°C, the 5-cell stack performance seems to be slightly improved, with higher current densities reached, most probably due to a better fluidic distribution on the short stack as compared to the 25-cell stack. Simultaneously, the gas manifold is also validated

Figure 5: initial iV curves in SOEC mode for G2 5-cell stack and 25-cell stack; 90%H₂O/10%H₂ with a flow rate of 12 Nml/min/cm² to the fuel electrode, air on the other side; a) plot of the voltages of each cell of each stack at 700°C; b) average cell voltage for each stack at 800, 750 and 700°C; steam conversion (SC) is provided on secondary x-axis

Stacks have also been operated in SOFC mode. Figure 6 presents the initial performances at 700°C in 100% H₂ for two flow rates for the 25-cell stack comprising G2 cells.
Figure 6: Initial iV curves in SOFC mode at 700°C for G2 25-cell stack; 100% H\textsubscript{2} with two flow rates of 1 and 3 NmL/min/cm\textsuperscript{2} provided to the fuel electrode, air on the other side; average cell voltage is reported; maximum fuel utilization is reported for each curve on the graph.

It can be seen that the stack could be operated in pure hydrogen up to very high fuel utilization, 100% and 81% for the two tested flow rates of 1 and 3 NmL/min/cm\textsuperscript{2}. The maximum current densities reached were 0.15 and 0.35 A/cm\textsuperscript{2} at 0.8 V in average per cell. These performances might look low as compared to those obtained on single cells and reported in figure 1, but it needs to be noticed that the flow rates are 3 to 10 times lower, which strongly affects the performances.

Durability tests:
Figure 7 presents long period rSOC load cycling test results for both REFLEX reference (so called ref) and REFLEX optimized (G2) 5 cells stacks. Cell voltages and current densities are plotted as function of test duration. Additionally, red and green dots indicate iV curves and the different iV curves are plotted in figure 8. Figure 7 a) and b) show respectively ref and G2 stacks results. Table 3 summarizes flow rate, current densities and corresponding reactant (fuel or steam) utilization for each stack.

Table 3: flow rate, current densities and corresponding reactant (fuel or steam) utilization for each testing phase of each stack

<table>
<thead>
<tr>
<th>Cell type</th>
<th>Mode</th>
<th>Plate</th>
<th>H2</th>
<th>H2O</th>
<th>i (A/cm\textsuperscript{2})</th>
<th>Reactant Utilization rate (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SOFC</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Ref</td>
<td>SOEC</td>
<td>EC# 1</td>
<td>6</td>
<td>6</td>
<td>-0.51</td>
<td>59</td>
</tr>
<tr>
<td>SOEC</td>
<td></td>
<td>EC# 2-1</td>
<td>6</td>
<td>6</td>
<td>-0.48</td>
<td>56</td>
</tr>
<tr>
<td>SOEC</td>
<td></td>
<td>EC# 2-2</td>
<td>6</td>
<td>6</td>
<td>-0.46</td>
<td>53</td>
</tr>
<tr>
<td>SOEC</td>
<td></td>
<td>EC# 3-1</td>
<td>6</td>
<td>6</td>
<td>-0.51</td>
<td>59</td>
</tr>
<tr>
<td>SOEC</td>
<td></td>
<td>EC# 3-2</td>
<td>6</td>
<td>6</td>
<td>-0.51</td>
<td>59</td>
</tr>
<tr>
<td>SOEC</td>
<td></td>
<td>EC# 4</td>
<td>6</td>
<td>6</td>
<td>-0.51</td>
<td>59</td>
</tr>
<tr>
<td>SOFC</td>
<td></td>
<td>FC#1 to FC#4</td>
<td>6</td>
<td>6</td>
<td>0.36</td>
<td>42</td>
</tr>
</tbody>
</table>

Current step duration was targeted to be 100 h to be performed four times for each mode to get 800 h of test (400h SOFC/ 400 h SOEC). Ref stack test drifted slightly from this objective, current steps were sometimes interrupted or extended over 100 h but finally
cumulated times are close for both stacks: respectively 506h/413h SOEC/SOFC for ref stack and 497h/402h for G2 stack.

**Figure 7-a**

**Figure 7-b**

**Figure 7**: Durability tests for reference and optimized (G2) 5 cells stack performed at temperature and flow rate reported in the figure.

For both tests, SOEC cell voltage increases during the operating time, with an increase of the voltage of 36 mV/1000h for ref cells and 17 mV/1000h for G2 cells. During the time periods of SOFC operation, voltage decreases of 12 mV/1000h for ref cells and 22 mV/1000h for G2 cells. As indicated above for initial performances, voltage scattering is higher for ref stack for which cell #5 performance was better than others (particularly in SOEC) over the duration of the test.

Figure 8 presents the iV curves recorded during the long period of rSOC load cycling test. Both SOEC and SOFC results are plotted. For a sake of clarity, some statistical treatment is performed: each point of the curves corresponds to the median of the cells voltages recorded at corresponding current density.
Figure 8: iV curves in SOEC and SOFC mode recorded during the rSOC test depicted in figure 7.

For each curve the legend sets out curve label and cumulated time for each mode (Fuel Cell FC or Electrolysis Cell EC) at iV curve recording time. Full diamond dot correspond to ref stack and empty circular dot to G2 stack. For both stacks iV curves scattering over time is significantly higher in SOEC mode. At very beginning of the test stack with G2 cells performed slightly better than ref cells stack but after completion of rSOC long period cycling, ref and G2 curves overlapped. Table 4 presents the ASR derived from the first and final curves for each stack. It shows an increase of 36% (ref) and 51% (G2), respectively, in EC mode. In FC mode the increase is significantly lower: 3% (ref) and 10% (G2).

Difference on ASR increase between ref and G2 could be partially explained by higher current density used for G2 test: -0.58 A/cm² vs. -0.51 A/cm² for ref in SOEC mode. According to this analysis, cell degradation in stack looks consistent with degradation observed at single cell test level.

Table 4: ASR derived from iV curves by linear regression of iV curves for i within -0.3 A/cm² and -0.1 A/cm² (SOEC mode) and i within 0.1 A/cm² and 0.3 A/cm² (SOFC) mode.

<table>
<thead>
<tr>
<th>Cell type</th>
<th>ID curve</th>
<th>ASR (µΩ/cm²)</th>
<th>Operating duration (h)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>SOEC</td>
<td>SOFC</td>
<td>SOEC</td>
</tr>
<tr>
<td>ref</td>
<td></td>
<td></td>
<td>SOEC</td>
</tr>
<tr>
<td>IV#1</td>
<td>0.540</td>
<td>0.450</td>
<td>0</td>
</tr>
<tr>
<td>IV#4</td>
<td>0.654</td>
<td>0.457</td>
<td>312</td>
</tr>
<tr>
<td>IV#7</td>
<td>0.736</td>
<td>0.462</td>
<td>506</td>
</tr>
<tr>
<td>G2</td>
<td></td>
<td></td>
<td>SOEC</td>
</tr>
<tr>
<td>IV#1</td>
<td>0.510</td>
<td>0.412</td>
<td>0</td>
</tr>
<tr>
<td>IV#7</td>
<td>0.690</td>
<td>0.447</td>
<td>331</td>
</tr>
<tr>
<td>IV#9</td>
<td>0.771</td>
<td>0.453</td>
<td>497</td>
</tr>
</tbody>
</table>

4. Conclusion

Based on the initial characterization via iV-curves on single cells (Figure 1) the targeted operation of the rSOC in the REFLEX project, that is 80% FU at 700°C, -1.2 A/cm² at 90% H₂O in H₂, and 0.6 A/cm² and dry H₂ in SOFC mode can be achieved but is on the limit for fuel starvation and leave the ALD cell, with a higher performance to start operating in SOEC mode below thermoneutral potential while the other cell (RND14) will start the SOEC operation above thermoneutral potential. The 250 h at constant SOEC operation at 700°C, -1.2 A/cm² at 90% H₂O in H₂ caused severe degradation ie. 790 mV/kh and 937 mV/kh, for ALD and RND14 respectively. Orders of magnitude lower degradation rates were observed both during constant SOFC operation (0.6 A/cm²) and reversible (load
cycling) operation. As a side result, it was noticed that the cell kept an OCV within 2 mV of the theoretical Nernst potential at H₂O/H₂:50/50; a good indicator that the only 2 µm thick electrolyte stays gas tight throughout the single cell tests depicted in Figure 2.

Cells of type RND14 (so-called G2 cells for REFLEX project) were produced at full scale and integrated into short stack for performance and durability test, and in full scale 25-cell stack. The initial performance of the stack comprising those cells is a little bit better than a stack made of reference cell, at 750 and 700°C. Scattering of voltages in the stack is smaller than for the stack made of reference cells. As for single cell tests, high steam conversion can be reached with a value close to 90% at 800°C for the flow rate considered. The same conclusion is obtained for the 25-cell stack, which fully validate the integration of G2 cells into the stack design considered for REFLEX project.

Regarding the durability test, it has been possible to apply slightly higher current densities in the stack made of G2 cells in both SOEC and SOFC mode, but lower than what could be achieved for the single cell. The stack, as the single cell, confirms a higher degradation rate in SOEC as compared to SOFC.

Acknowledgement

The authors would like to thank the project REFLEX for financial support. REFLEX is funded by the Fuel Cells and Hydrogen 2 Joint Undertaking under the European Union’s Horizon 2020 research and innovation program under grant agreement No 779577. The authors also thanks colleagues at DTU Energy (Technical University of Denmark) for technical assistance and valuable discussions, and at CEA for stack design, manufacturing and testing.

References