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Stack Optimization and Testing for its Integration in a rSOC-Based Renewable Energy Storage System

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Stacks dedicated to rSOC operation require improvements as compared to stacks dedicated to purely SOEC or SOFC mode. Starting from an electrolysis stack, improvements have been performed in the European project REFLEX, mainly to enhance reactants distribution, reduce pressure drops, integrate new cells specifically developed as part of REFLEX project, and finally integrate larger cells to reduce stack and system cost and footprint. For easier handling, mechanical connection to the system was optimized.

Long-term degradation tests were performed both for reference and optimized cells within two 5-cell stacks. A full size 25-cell stack was assembled integrating optimized connections to gases lines, specific stack clamping system and internal electrical insulation required for stack integration into REFLEX modules.

For prospective reason enlarged cells were produced and integrated within first a 5-cell stack, and then a 25-cell stack. Finally, stability of performance along pre-serial manufacturing process was checked for 20 stacks before their delivery for integration into REFLEX modules.

Introduction

Solid oxide cell (SOC) technology has the advantage to operate as a “fuel versatile” fuel cell (SOFC) oxidizing directly H\textsubscript{2} or CO (1, 2) but also, indirectly, fuels as NH\textsubscript{3} or CH\textsubscript{4} (3, 4). Moreover, the same cell can be operated in electrolysis mode producing H\textsubscript{2}, CO or syngas (5-8). Obviously, those cells can be operated as reversible solid oxide cells (rSOC) offering the possibility of long-term bulk energy storage (9-13): H\textsubscript{2} produced using rSOC as SOEC (electrolysis mode) in case of excessive electricity production, can subsequently be oxidized within the same system to supply electricity. Separate ‘power-to-gas’ and ‘gas-to-power’ components are not needed which should reduce costs. Sylfen Smart Energy Hub\textsuperscript{TM} (SEH) concept constitute an instance of hybrid energy storage (both through battery use and H\textsubscript{2} production) and cogeneration system to provide energy to buildings and eco-districts. The concept was proved by SmartHyes demonstrator (14), developed for ENGIE using one CEA Stack (15) as core of the rSOC. Smart Energy Hub\textsuperscript{TM} development is currently performed in the frame of European project REFLEX\textsuperscript{1} (16). REFLEX goal is to operate in-field SEH first prototype managing three rSOC modules of four stacks each that

\textsuperscript{1} Reversible solid oxide Electrolyze and Fuel cell for optimized Local Energy mix
will be installed (2021) at Envipark in Torino and coupled to PV local field and mini-hydro power plant to provide electricity and heat to the headquarter of the park.

To reach the high power-to-power (P2P) round-trip efficiency REFLEX goal, developments were achieved at system level, to increase power conversion and storage subsystem efficiency, to design, by mean of numerical simulations, a compact and thermally efficient architecture and to develop a smart and flexible in-field system management application. Increase in performances, in both SOFC and SOEC modes, was also expected from the electrochemical core of the system acting at cell as well as at stack levels. In fact, rSOC cells and stacks have to cope with harsh operating conditions in that sense that they are exposed to rapid voltage inversion and gas composition changes but also alternation between exothermic, adiabatic, and endothermic states. Moreover, it is obvious that, to reach high efficiency level, reactant utilization rate has to be as high as possible involving the necessity for flow rates to be uniformly distributed over each cell active area. That is the only way to limit the risk of local shortage of reactant that could lead to irreversible cell degradation and finally drastic system performance drop or shutdown if stack integrity is compromised. CEA stack as described in (15), was developed to operate specifically in SOEC mode and had to be optimized for SOFC mode at the start of REFLEX project. Optimized cell (so called G2) was specifically developed (17) from a standard Elcogen cell (so called reference or Ref cell). Stack optimization process was so initiated by fabricating a stack comprising five Ref. cells as well as five-cell stack comprising G2 cells to investigate, by comparison, durability at load cycling operation as performed at cell level (18). To facilitate mechanical handling during SEH integration process the fitting of the distribution and collection pipes of gases to the stack was improved. Additionally, an electrical insulation was created between electrochemical cells and terminal flanges. Floating stacks now enable series connection. These operational optimizations as well as stack clamping system (19) were validated by fabricating a full size (25-cells), first of its kind, stack comprising G2 cells. Active area of cell was extended and tested at both scales, five and 25 cells stacks. Finally stacks to be implemented in the REFLEX SEH were produced and qualified in terms of initial performances.

**Experimental**

**Cell specifications**

The reference structure for an Elcogen cell (so-called 400-B-SM, or REF cell) is a fuel electrode supported cell, consisting of ~380 µm Ni/3YSZ support layer, a ~5 µm Ni fuel contact layer, a ~12 µm Ni/8YSZ fuel active layer, a ~7 µm thick LSC oxygen electrode, a ~2µm 8YSZ electrolyte and a CGO barrier layer of similar thickness.

In the frame of REFLEX project this structure was optimized in so-called G2 cells as previously detailed (17).

Cells used to manufacture stacks needed during optimization process were square shaped. For first optimization steps that led to obtain the first of its kind 25 cells REFLEX stack, cell size was 120×120 mm², with a squared shape active area of 100 cm². The same cell size is considered for the stacks to be included in the SEH modules.

Enlarged cells structure was similar to Ref. cell and its size is 160×160 mm² with a squared shape active area of 196 cm².
**Stack specification**

At the start of optimization process, CEA stack corresponded to the description given in (15): it was based on thin interconnects using 0.2 mm AISI441 ferritic stainless steel sheets. It comprised almost 500µm thick cells from Elcogen of 100 cm² in active area. A nickel-mesh and an LSM contact element were set in the H₂ and O₂ compartments respectively. A cross flow design was chosen. Sealing was achieved with a commercial ceramic glass. A mica foil was added to ensure the electrical insulation between two adjacent interconnects, but also to complete the sealing and to precisely position the cell. To optimize the sealing and the electrical contact, a mechanical loading was applied on the stack by external system. Thermocouples (K-type) were located in holes drilled in the thick end plates of the stacks, accuracy at test temperature is ±3°C. Current was applied with two current rods fixed to the end plates. Voltage probes were spot-welded to each interconnect to measure the voltage of each cell.

Distribution of reactants and collection of reaction products were performed by mean of four tubes, two for fuel side and two for air side. This was convenient for laboratory reasons because doing so the stack could be easily connected to any test bench. During the optimization process, stack assembly remained quite similar even if some dimensions had to be adjusted to integrate the thinner and larger cells. The connection to the reactants and reaction products lines has also been improved to be more industrial.

**Test benches**

Stacks tests reported in the present paper were conducted on multiple benches as stacks as well as cell sizes were enlarged during the testing period. Instrumentation system was very similar for all of them. The stack was connected both to a power supply and to an electronic load for rSOC operation. Current and stack voltage are recorded as well as individual cell voltage. Hot wires mass flow-controllers adjusted the gases supply and mass flow meters were set at the outlet of each compartment, after condensation of the unused water in case of the hydrogen side, in order to evaluate the stack gas tightness at Open Cell Voltage (OCV) or under polarization.

Accuracy of the multiple instrumentation systems that equip the benches is in the same order of magnitude. Main measurement accuracy specifications are the following: current ± 0.5 A, total stack voltage ±0.18 V, individual cell voltage ±10mV, pressures ±5mbar (at worst), mass flows ±3%.

Counter pressure force needed to ensure mechanical coherence of the stack was applied by electric jack for load cycling tests and enlarged cells stack tests, while self-clamping system (19) was used for the stacks to be implemented within REFLEX Smart Energy Hub.

**Electrochemical test**

For each stack produced and tested through the optimization process, the testing procedure started by heating the stack for sealing and reduction. After reduction, the stacks were initially characterized by polarization (so called i-V) curves in both mode at 800°C, 750°C, and 700°C.

For durability tests, 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, the stack was supplied with a mixture of 50/50 vol.% H₂O/H₂ at total flow rate of 12.0 NmL min⁻¹ cm⁻² on fuel side and air (clean and dry) on oxygen side. Stack performances were checked at each change of operating mode through i-V curves recorded at 800, 750 and 700°C. At first stage of the test, less performing cell voltage was adjusted at 0.8V in SOFC mode and 1.3 V in SOEC. Then the test was operated in galvanostatic mode. At each change, a set of i-
V curves corresponding to the initial one was recorded. A second stage of more than 250h of operation consisted to alternate daily from SOEC to SOFC by cycles. For each testing day SOFC step was targeted to be 16h at 0.3 A/cm² (fuel utilization FU 70%) and SOEC step 8h at -1.2A/cm² (steam conversion SC 77%). 20 cycles were targeted to get 480h in operation at least.

During this phase gas compositions were adjusted: for SOEC steps feeding gas at H₂ electrode was 90/10 vol.% H₂O/H₂ at total flow rate of 12.0 NmL min⁻¹ cm⁻², O₂ electrode was fed with dry air to control the internal differential pressure. During SOFC steps stacks were fed by flow of 3.0 NmL min⁻¹ cm⁻² of dry H₂ on fuel side, and 5.4 NmL min⁻¹ cm⁻² of dry air on air side. Degradation was evaluated both with the evolution of voltage over time and with the ASR obtained from iV curves recorded before and after each step of the test.

Results and Discussion

Fluidic optimization of the stack

The level of performance expected at stack level to meet REFLEX project requirements involved increasing the flow of reactants. So, both internal gas path and mechanical connections to the reactant and reaction products lines had to be improved to limit pressure drops. A special work was done as reported in (16) on internal air path because CEA stack, initially developed to operate specifically in SOEC mode, must now cope with high airflow rates required for SOFC mode. It was observed (16) at Single Repeat Unit scale the pressure drop was reduced by a factor higher than two.

To remove the connection tubes, described above, some mechanical connections based on flange and metallic gasket solution have been developed. Stack can now be easily connected to a manifold plate, part of the system, ensuring gas distribution and collection functions as well as mechanical support of the stack. This improvement was implemented on enlarged cells stacks for testing but also on stacks produced for REFLEX modules.

Load cycling tests

Results of long period load cycling have already been presented (20), the main one being that even though it was operated under higher current density -0.58 A cm⁻² versus -0.51 A cm⁻² during the SOEC steps, totalling a duration of 400 h, the degradation of the G2 cell is very comparable to that of the ref cell as shown by iV curves, recorded at the end of the tests which strictly overlap, (see Fig 8 (20)).

Figure 1 presents the results of daily switching rSOC test, respectively for stack comprising five Ref cells (Figure 1-a) and stack comprising five cells G2 (Figure 1-b).

Actual tests were not completely conducted as initially planned due to different incidents. Table 5 summarizes operating conditions for both stacks.

- For ref stack: 18 day cycles were performed, some of them do not reached duration targets, the last 10 cycles were more regular.
  - Cells degradation is not so clear in SOEC mode, for cell #5 an increase of 32 mV is observed during the test (≈ 5%/kh) but the cell #1 voltage remains constant over the test, in SOFC mode, voltage reduction stay within 10 to 15 mV and so degradation rate evaluated within the range of 2 to 4 % kh⁻¹.

- For G2, only ten daily cycles were done which were closer to the objective in term of regularity. Unfortunately, this test was performed after unexpected thermal cycle that impacted cells performance. Consequently operating conditions were milder
than for ref. Despite this, observed degradation is quite similar to ref: in SOEC for the best performing cell (cell #4) voltage increased by 20 mV (degradation rate ≈ 6% kh\(^{-1}\)) but not significant change was observed for the worst performing cells (#1&2). In SOFC voltage losses remain within 5 to 10 mV (degradation rate ≈ 2 to 4% kh\(^{-1}\)).

It seems noticeable that during SOEC steps, for all reference cells, voltage decreases and that this behavior is observed for two cells of G2 stack as well, concurrently with an increase of voltage for the three others. That could be due to some thermal effect, cells operated above thermoneutral voltage having voltage decreasing over time, while cells being operated below thermoneutral voltage having voltage increasing over time.

To be more conclusive this kind of test should probably have to be optimized including, at least, more instrumentation, to track thermal effects for example.

![Figure 1: Daily switching tests for: a) Ref cell and b) G2 5-cell stacks, performed at 700°C. During 16h SOFC steps, stacks were fed with dry pure H\(_2\) flow rate of 3 NmL min\(^{-1}\)cm\(^{-2}\) on fuel side and 5.4 NmL min\(^{-1}\)cm\(^{-2}\) on air side. For 8h SOEC steps stacks were fed with 90/10 vol.% H\(_2\)O/H\(_2\) mix at total flow rate 12 NmL min\(^{-1}\)cm\(^{-2}\) on fuel side and dry air on air side to ensure required differential pressure between gas compartments.](image)


TABLE I. Operating conditions for short stacks during daily switching rSOC test.

<table>
<thead>
<tr>
<th>Cell ID</th>
<th>Mode</th>
<th>Flow rate / NmL min$^{-1}$ cm$^{-2}$</th>
<th>Current density / A cm$^{-2}$</th>
<th>Reactant Utilization / %</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ref</td>
<td>SOEC</td>
<td>1.2 10.8</td>
<td>-0.84</td>
<td>54</td>
</tr>
<tr>
<td>SOFC</td>
<td></td>
<td>3.0 0</td>
<td>0.28</td>
<td>65</td>
</tr>
<tr>
<td>G2</td>
<td>SOEC</td>
<td>1.2 10.8</td>
<td>-0.44</td>
<td>28</td>
</tr>
<tr>
<td>SOFC</td>
<td></td>
<td>3.0 0</td>
<td>0.18</td>
<td>42</td>
</tr>
</tbody>
</table>

Stack comprising enlarged cells

Figure 2 presents iV curves recorded at 800°C for a 5-cell stack made of enlarged cells. All the cells of the stack were instrumented, but for reason of picture clarity only three curves indicating at each current density value, respectively the minimum, median and maximum voltage observed on cells are plotted and compared to the same type of plot for a 5-cell stack comprising 100 cm$^2$ ref cells. Voltage level evolution as current density increases is very comparable for both stacks. Voltage scattering is, also, quite similar for reference and enlarged 5-cell stack. Steam starvation effect appears at the same level of steam conversion slightly higher than 80%, which shows that even on enlarged cells fluidic distribution is fine.

Figure 2: Initial iV curves in SOEC mode at 800°C for five ref cell stacks respectively 100 cm$^2$ of active area, red curves, and 196 cm$^2$ of active area, blue curves. Total flow rate of 12 NmL min$^{-1}$ cm$^{-2}$ of 90/10 vol.% H$_2$O/H$_2$ mix is provided to the fuel electrode, air on the other side. Minimum, median and maximum cell voltages are represented by short dotted lines, full lines and long dotted lines respectively. Steam conversion is reported by secondary abscissa axis on top of the graph.

The curves show that cell enlargement has no negative impact on stack performance, median cell voltage is even lower, and validates the integration of enlarged cells in the stack, at 5-cell stack scale, first.

A 25-cell 196 cm$^2$ active area stack was fabricated equipped with the new mechanical connections of the gas distribution and collection lines, shortly described above, and with the internal electrical insulation necessary for the operation of the REFLEX modules based on two stacks connected in series. Figure 3 presents the iV curves recorded on this stack in
SOEC operation and compares them to those of the 5-cell stack fabricated with enlarged cells presented in figure 2, but also with a “classical” stack comprising 25 cells of 100 cm² of active area. It shows that performances at 25 cells scale is close to those already observed for 5 enlarged cells stacks. Discrepancies can be partially explained by thermal effect, since temperature is a little bit less than 800 °C (796°C) at iV curve recording start and decreased, about 6°C, during the iV curve recording.

Cells performance scattering, about 100 mV at -1.20 A/cm², is consistent with the previous observations.

The curve of the 25 enlarged and 25 ref cells stack almost overlap. It validates the integration of enlarged cells in a full scale stack, from a mechanical and fluidic point of view since performances are similar.

![Graph showing initial iV curves in SOEC mode at 800°C for ref cells stacks, respectively 25 cells of 100 cm² active area: green circle dots, five cells of 196 cm² active area: blue square dots and 25 cells of 196 cm² active area: yellow diamond dots. Total flow rate of 12 NmL min⁻¹ cm⁻² of 90/10 vol.% H₂O/H₂ mix is provided to the fuel electrode, air on the other side; median cell voltage is reported for each stack; steam conversion is reported by secondary abscissa axis top of the graph.](image)

**Figure 3:** Initial iV curves in SOEC mode at 800°C for ref cells stacks, respectively 25 cells of 100 cm² active area: green circle dots, five cells of 196 cm² active area: blue square dots and 25 cells of 196 cm² active area: yellow diamond dots. Total flow rate of 12 NmL min⁻¹ cm⁻² of 90/10 vol.% H₂O/H₂ mix is provided to the fuel electrode, air on the other side; median cell voltage is reported for each stack; steam conversion is reported by secondary abscissa axis top of the graph.

**Stacks production for REFLEX demonstrator:**

REFLEX SEH demonstrator will manage three modules each comprising four stacks. The CEA has therefore manufactured twelve stacks plus four spares for the field tests but also four additional stacks for preliminary tests in Sylfen laboratory.

Stack production and delivery were organized by batches of four stacks.

- **Batch 0,** is the first set of stacks delivered to Sylfen in order to perform tests of system architecture, auxiliaries performance and system control at Sylfen laboratory. Stacks of this first batch, comprising ref cells, do not include electrical insulation that enables electrical series connection.
- **Batch 1 to 3** will be used in REFLEX demonstrator rSOC modules to be operated at Envipark (Torino). All the stacks have an internal electrical insulation so that they can be connected in series. These stacks are equipped with G2 cells of 100 cm² of active area.
- **Batch 4** is a spare and so of the same kind as Batches 1 to 3, except that two of them are comprising ref cells.
Figure 4 (a to c) shows the performance iV curves recorded after stack manufacturing and conditioning processes. The curves highlights that stacks behavior (symbolized by the median of cells voltage of each stack for reason of picture clarity) is quite similar from one stack to another, especially for the REFLEX optimized stacks (Batch 1 to 3).

Finally, one has to notice that the scattering of stacks median cell voltages within the total production is in the same order of magnitude as the scattering of cells voltage observed internally in the stack (see Figure 5).

All the stacks were not assembled on the same bench, and because this new bench could not be operated in SOEC mode, SOEC curves of spare stacks are missing on Figure 4-b&c. Nevertheless, Figure 4-a recorded in SOFC mode shows that the stacks manufactured for final tests of REFLEX have close performances.

Figure 4: Acceptance iV curves of REFLEX stacks five batches of four stacks each, all performed at 700°C. Fig 4-a: stacks operated in SOFC mode. Flow rate of 1 NmL min\(^{-1}\) cm\(^{-2}\) of pure H\(_2\) is provided to fuel electrode, air on the other side. Fig 4-b&c stacks operated in SOEC providing 90/10 vol.% H\(_2\)O/H\(_2\) mix to the fuel electrode, air on the other side, total fuel flow rate 12 and 6 NmL min\(^{-1}\) cm\(^{-2}\) respectively. For each curve, median cell voltage is reported for each stack, steam conversion is reported by secondary abscissa axis top of the graph. Color of the line identify manufacturing batches: blue for batch 0 delivered to Sylfen for in-lab tests of the SEH, green, yellow, red for batches 1, 2 and 3 to be implemented within REFLEX SEH modules, and finally black for spare batch 4.
Figure 5: Cell voltage typical scattering recorded for a stack operated in SOEC providing 90/10 vol.% H₂O/H₂ mix to the fuel electrode at 700°C. Cells voltage data are plotted with circle marks, the associated median cell voltage is plotted as a red continuous line, two dotted lines derived from median voltage curve highlight cell voltage scattering remains within ±7%.

**Conclusion**

As part of REFLEX project that aims to demonstrate in-field the efficiency of a rSOC based renewable energies storage, system designed by Sylfen, a specific task was to optimize CEA stack design.

First it was necessary to adapt the stack concept initially devoted to hydrogen production purposes to operate as a performing rSOC device. Areas of improvement identified were (i) the modification of internal gases paths to limit pressure drop and optimize the uniformity of cell feeding particularly on the air side, that was not an issue in pure SOEC mode (ii) the integration within the stack of new cells specifically developed for rSOC operation as a project target (iii) the improvement of mechanical connections to distribution of reactants and collection of reaction products lines to facilitate stack maintenance and handling in the field, (iv) the integration of a specific stand alone system for clamping and transport (v) to make the stack electrically floating by internal electrical insulation adjunction in order to electrically connect stacks in series within REFLEX modules.

Step by step these developments were integrated to the CEA stack and validated by specific tests. Particularly the REFLEX new cells were integrated in a five cells stack that was operated hundreds of hours in load cycling test.

Enlarged cells with a nearly doubled active area to reach 196 cm² were also produced by ELCOGEN, integrated within 25 cells CEA stack for which performances were found similar to 25 cells of 100 cm² stack despite the requirement of higher total gas flow and upscaling of mechanical parts.

Finally, twenty stacks were produced for REFLEX project: four for Sylfen laboratory validation purposes, twelve to be implemented within REFLEX modules and four as spare. The acceptance tests of the production demonstrate that even though it remains quite handcrafted, at this stage, the CEA stack production process leads to acceptable scattering in stacks properties.
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