# DELIVERABLE REPORT

# D3.3 DEFINITION OF LONG TERM STACK TEST PROTOCOL

GrInHy2.0
Green Industrial Hydrogen

Green Industrial Hydrogen via steam electrolysis


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## Table of acronyms

| | |
|---|---|
| ASR | Area Specific Resistance |
| BoL | Beginning of Life (of the stack) |
| CEA | Commissariat à l'Energie Atomique et aux Energies Alternatives |
| EIS | Electrochemical Impedance Spectroscopy |
| EoL | End of Life (of the stack) |
| OCV | Open Circuit Voltage |
| RES | Renewable Energy Sources |
| SC | Steam Conversion |
| SF | Sunfire |
| StE | Steam Electrolysis |
| TC | Thermocouple |

# 1 Introduction

The European Commission and its roadmap for moving towards a competitive low-carbon economy in 2050 sets greenhouse gas emissions targets for different economic sectors. One of the main challenges of transforming Europe´s economy will be the integration of highly volatile renewable energy sources (RES). Especially hydrogen produced from RES will have a major part in decarbonizing the industry, transport and energy sector – as feedstock, fuel and/or energy storage.

Access to renewable electricity will also be a limiting factor in the future and energy efficient technologies the key. Due to a significant energy input in form of steam preferably from industrial waste heat, Steam Electrolysis (StE) based on Solid Oxide Electrolysis Cells (SOEC) achieves outstanding electrical efficiencies of up to $84\%_{el,LHV}$. Thus, StE is a very promising technology to produce hydrogen and oxygen most energy efficiently.

In context with the production of green hydrogen from a steam electrolyzer, the steel industry combines both hydrogen and oxygen demand – today and future – and the availability of cost-efficient waste heat from its high-temperature production processes. Currently, the $H_2$ demand is mostly met using natural gas, and as a consequence is generating $CO_2$. Switching the source of $H_2$ from fossil fuel to StE using RES would naturally allow reducing greenhouse gas emissions, while favoring the development of a highly innovative and energy efficient technology. This context constitutes the frame for the EU project GrInHy2.0, the continuation of the first project that ran from March 2016 to March 2019, and is summarized in the following Figure 1:

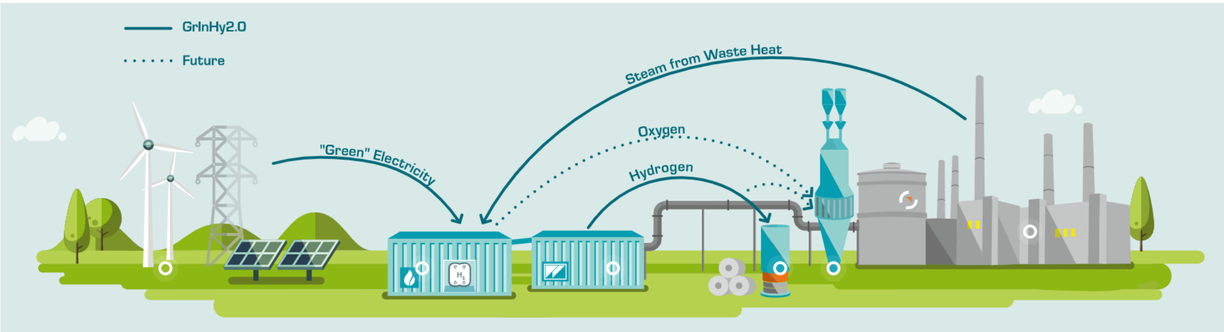



*Figure 1: Context of the GrInHy2.0 EU project.*

However, for an efficient use of StE in the steel industry, or any industry, the demonstration of on demand availability of the $H_2$ production must been made over significant duration. In this frame, one of the objectives of the GrInHy2.0 project is the demonstration of 20,000 h of operation, or 20 kh, of the Sunfire StE technology in stationary conditions. This amounts to approximately 2 years and 3 months of continuous operation, and will be carried out by CEA. This public deliverable is aimed at defining the experimental conditions related to this Task 3.4. It includes operational parameters, as well as nominal and emergency procedures. Finally, it details the initial, long term, and final test sequences built around the 20 kh targeted duration of the durability test.

## 2 Objectives of the Task

There are multiple objectives for performing this very long term durability testing. The main one, as mentioned before, is the demonstration that the technology can be operated over such durations. Indeed, longer life expectancy of stack means lower $H_2$ cost, since stack replacement remains a major economic factor. A key parameter in degradation studies is the degradation rate, which can be expressed in terms of voltage increase over time ($+mV.kh^{-1}$), or in terms of an increase of the area specific resistance over time ($+m\Omega.cm^2.kh^{-1}$).

Then, degradation mechanisms are still not perfectly understood, as they depend on cell and stack architectures, choice of materials, operating conditions, etc. The identification of the main sources of degradation should in turn allow extending the lifespan of the stacks.

Finally, meaningful knowledge is expected to be acquired along the way in the field of stack control, failure prevention and large data analysis, for example.

## 3 Operational Parameters

The targeted operating conditions for the 20 kh durability test are gathered in the following Table 1. Due to this report being public, some crucial information has been redacted to protect the intellectual property of the consortium.

The targeted Steam Conversion (SC) of 70% is fairly high. This represents the proportion of steam electrochemically turned into $H_2$ compared to the amount of steam actually fed to the stack inlet. High SC values have a direct and positive impact on the final cost of $H_2$ production.

*Table 1: Targeted operating conditions for the durability test*

| Variable | Value | *Units* | Comment |
|---|---|---|---|
| Current density | -0.65 | *$A.cm^{-2}$* | |
| SC | 70 | *%* | |
| $H_2O$ flowrate | 1192 | *$g.h^{-1}$* | |
| | 24.8 | *$NL.min^{-1}$* | |
| $H_2$ flowrate | 2.8 | *$NL.min^{-1}$* | |
| Air flowrate | 40 | *$NL.min^{-1}$* | |
| $pH_2O$ | 0.90 | *Atm* | |
| $pH_2$ | 0.10 | *Atm* | |
| $T_{STACK}$ | $T_{BoL}$ | *°C* | At Beginning of Life (BoL) |

# 4 Experimental Procedures

This paragraph describes nominal startup and shutdown procedures, as well as all procedures related to data recording.

The stack will be received at CEA with the fuel electrodes already in a reduced state. No NiO reduction procedure is thus given in this document.

## 4.1 Nominal Startup Procedure

The stack will be brought up to the operating temperature in air and a $H_2/N_2$ mixture. As long as the stack temperature is below 600 °C, which is the auto-ignition temperature of $H_2$ in air, the amount of $H_2$ in $N_2$ will be limited to 3 vol% for safety reasons. Indeed, at ambient temperature, mixtures with ≥4 vol% $H_2$ in air can explode. Above 600 °C, the proportion of $H_2$ will be increased to reduce the risks of cermet oxidation.

Once the targeted temperature is reached and the stack thermal response is stable, the composition will be switched before ramping up the DC current.

## 4.2 Nominal Shutdown Procedure

The nominal shutdown procedure is composed of same steps as the startup procedure, described in the above paragraph, in reversed order.

## 4.3 Stand-by Conditions

In case of a need to leave the stack in temperature at OCV, the following standby gas conditions will be used, depending on the duration of the idle period:

- Short Idle Time:

The stack will be left at OCV and nominal humidified gas conditions.

- Long Idle Time:

If the stack has to be left idle for a longer period of time (e.g. >12 h), the gas conditions will be switched to the one used in the nominal startup and shutdown procedures.

## 4.4 Recording of Performances

The performances of the stack, or iV characteristics, will be recorded over the useful range of temperature. The objective here will be to experimentally establish the strong relationship between individual cell performance and temperature.

For characterizing the stack performance, different approaches will be tested and evaluated. One methodology adopted to record the performances will be in accordance with the recommendations of the SOCTESQA project that can be found at:

http://www.soctesqa.eu/test-modules/tm03a-jv-short-decription.pdf

The following Figure 2 gives an example of what can be expected when recording a performance curve with this method.

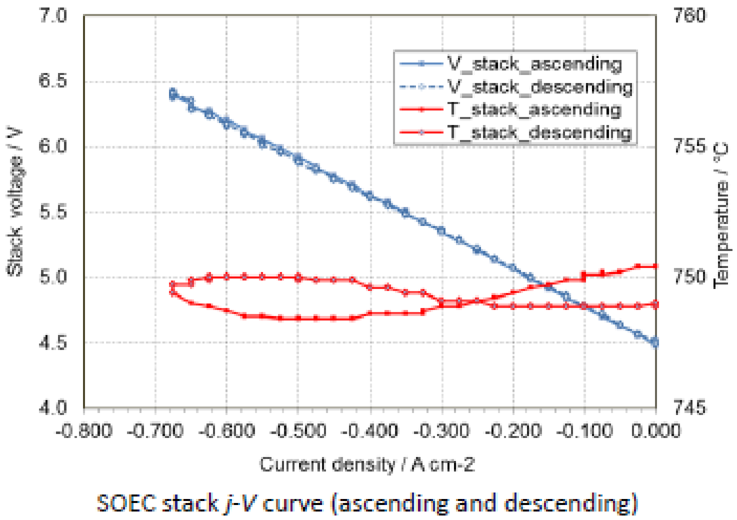



*Figure 2: Example of iV curve recording, taken from the corresponding module of the SOCTESQA project.*

The applicability of this methodology to a SF stack will be evaluated and the results will be compared to the polarization curves described in the following paragraph.

## 4.5 Recording of Iso-SC Polarization and Thermoneutral Curves

The iso-steam conversion polarization curve consists in recording multiple steady states in which the SC is equal to the target value. Therefore, all data points correspond to different flowrates. For the GrInHy2.0 project, the SC of interest is 70%, which should put the stack near the thermoneutral voltage over the range of flowrates considered.

Such a curve is interesting, especially from a system perspective, since all points along the recorded curve are potentially viable long term operating points.

The targeted operating conditions are gathered in the following Table 2. A particular attention will be given the thermal stability of the complete stack response before recording.

*Table 2: Experimental conditions for the recording of the iso-SC polarization curve. The furnace temperature will be adjusted to maintain the stack in a near thermal-neutral state.*

| **SC** | **$T_{stack}$** | **i** | **I** | **$pH_2O$** | **$pH_2$** | **$F(H_2O)$** | | **$F(H_2)$** | **F(Air)** | **$F_c$** | **$F_a$** | **$F_a/F_c$** |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| *%* | *°C* | *$A.cm^{-2}$* | *A* | - | - | *$g.h^{-1}$* | *$NL.min^{-1}$* | | | *$NmL.min^{-1}.cm^{-2}$* | | - |
| 70 | $T_{BoL}$ | -0.20 | 25.6 | 0.90 | 0.10 | 367 | 7.6 | 0.8 | 12.3 | 2.2 | 3.2 | 1.45 |
| | | -0.35 | 44.7 | | | 642 | 13.3 | 1.5 | 21.4 | 3.9 | 5.6 | |
| | | -0.50 | 63.9 | | | 917 | 19.0 | 2.1 | 30.6 | 5.5 | 8.0 | |
| | | -0.65 | 83.1 | | | 1192 | 24.8 | 2.8 | 40 | 7.2 | 10.4 | |

Another characteristic curve will be recorded at a full thermal neutral state, i.e. at different temperatures.

## 4.6 Electrochemical Tightness Test

This test will aim to quantify the tightness of the stack. The procedure consists in filling the stack with $H_2$ on the fuel side, and air on the air side, before switching off gas flowrates and recording the evolution of the open circuit voltages (OCV) of the cells over time. A particular attention will be given to making sure that the complete setup is dry, and has been purged from all traces of steam. At the end of the test, the gas supply will be reinstated.

The quality of this tightness test will be assessed by comparing the results to previous ones obtained at CEA and SF, and comparing all tests to each other.

This test should be run at BoL and EoL, as well as after all unplanned interruptions and scheduled electrical shutdowns.

## 4.7 Electrochemical Impedance Spectroscopy

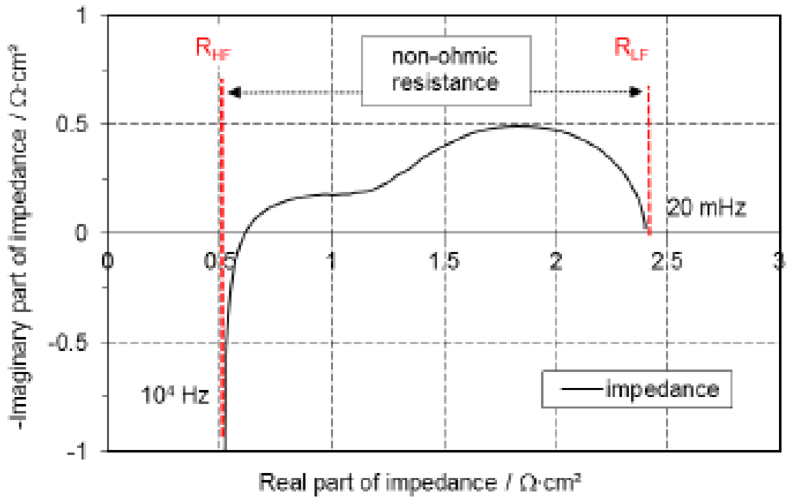
Electrochemical Impedance Spectroscopy (EIS) is a powerful analysis tool that can help study the response of the stack and its evolution over time. It consists in the recording of an oscillating voltage response in response to an oscillating current stimuli. The frequency of the stimuli is varied over large range, and the analysis of the impedance of the system as a function of the frequency typically allows identifying elementary mechanisms as well as quantifying the degradation over time.

In order to record high quality data, 4 wires are needed: 2 to supply the current, and 2 to measure the voltage response. The optimum position of for all wires will be determined during the preliminary testing phase.

In the frame of the GrInHy2.0 project, the recording of EIS spectra will be done in accordance with SOCTESQA project recommendation that can be found at:

http://www.soctesqa.eu/test-modules/tm04a-eis-short-description.pdf

The following *Figure 3* gives an example of EIS spectrum that can be recorded on Solid Oxide Cells.




Cole-Cole plot representing imaginary part vs real part of impedance

*Figure 3: Example of EIS spectrum, taken from the corresponding module of the SOCTESQA project.*

# 5 Preliminary Testing

The complete test bench and its modifications for the purpose of the GrInHy2.0 project will first be validated using a dedicated stack. Particular attention will be devoted to the gas supply lines and the manifold, as this last element has been manufactured by CEA based on Sunfire drawings and dimensions.

The furnace that will be used for the tests is equipped with 3 heating zones. The first 2 will heat the top and bottom part of the stack, respectively. The third one will make sure that gases reach the stack at the desired temperature. The preliminary phase will be used to adjust if needed the control parameters (PID) of the different heating elements to secure an appropriate level of temperature control.

Finally, the objectives of the preliminary testing are to determine the best method for the characterization of the stack performance and the best wire configuration for EIS measurements. For the purpose of the latter, 3 cells located at the top, center, and bottom of the dedicated stack have been equipped with multiple voltage sensors each. All possible configurations will be tested to determine the optimum, and the results will be applied to equip the stack destined to be operated 20 kh with appropriate sensors.

# 6 Testing Protocol

## 6.1 Initial Performance Test

The stack will be brought to the operating temperature and gas conditions following the nominal startup procedure. Then, after checking the tightness of the complete setup, and the stack in particular, the BoL performance and EIS data will be recorded over the appropriate range of temperatures. The proper durability test in the conditions described in Section 3 will then be started.

## 6.2 Long Term Test

The targeted duration for the long term test is 20 kh. Every 2000 h, the stack will be brought back to zero polarization (OCV) to record impedance data and assess the degradation. After 10 kh, or mid-way through the complete test, the performances will also be recorded in addition to the typical EIS data.

After every thermal cycles, and particularly after each annual electrical shutdown, the electrochemical tightness of the stack will be evaluated.

## 6.3 Final Performance Test

At the end of the test, if the state of the stack allows it, the performances, the iso-SC polarization curve and the thermos neutral curve will be recorded, in addition to the EIS data. The electrochemical tightness test will also be run before cooling off the stack.