Design, laboratory characterization, factory acceptance tests and installation of a 2.6 MW High temperature steam electrolyser in a refinery

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ABSTRACT

High-temperature steam electrolysis (HTSE) is the most efficient technology to produce hydrogen. MULTIPLHY European project aims to install and operate a HTSE system in multi-megawatt-scale (~2.6 MW), at a renewable products refinery. The HTSE unit will produce more than 60 kg_{H2}/h. It is complemented by a Hydrogen Processing Unit to meet the quality and pressure criteria of the refinery process. Both are installed on NESTE Rotterdam refinery, where the site has been prepared to receive them and to provide the requested power line and other utilities. Onsite installation and commissioning have been performed. In parallel, long term stack tests have been performed at laboratory scale in order to assess the technology durability. Stacks of power up to 19 kW have been tested over durations up to 8 kh. Thanks to an operation strategy consisting in compensating degradation by a stack temperature evolution over time, they could be operated over those durations without any hydrogen production loss, the current density being kept constant over the whole duration.

1. Introduction

Currently, 95 million tons (Mt) of hydrogen are used globally [1], with 8.2 Mt consumed in the European Union (EU) [2], primarily for industrial applications such as refining, ammonia production for fertilizers, and other chemical processes [1,2]. The majority of this hydrogen is currently produced from fossil fuels, 62% via steam methane reforming and 21% through coal gasification [1], emitting 11 and 20 kg_{CO2} per kg of hydrogen, respectively [3]. The remainder comes from by-product hydrogen generated at refineries and in the petrochemical sector. As a result, there is a need to develop alternative production methods that emit significantly less CO_2. Looking ahead, it is expected that these cleaner production technologies will replace fossil-based hydrogen, helping to decarbonize these industries. Beyond its traditional industrial uses, hydrogen is anticipated to play a key role in new applications within the context of the energy transition. Within the industrial sector, hydrogen could be used, either directly or indirectly, to decarbonize "hard-to-abate" sectors. For example, in the steel industry, hydrogen can be used directly as a substitute for coke in the iron

reduction process. It can also be used indirectly to generate high-grade heat in burners, replacing natural gas in industries like cement, glass, or metallurgy. Hydrogen is also being considered as a fuel for sectors where batteries are not feasible, such as heavy-duty or long-distance transport (e.g., trucks, ships, or aircraft), either through direct use in fuel cells or engines/turbines, or indirectly to produce e-fuels when combined with CO_2. Lastly, when produced via electrolysis, hydrogen offers a means to store renewable electricity and enhance the stability and flexibility of the electrical grid.

Clean H_2 produced via water electrolysis from Renewable Electricity Sources (RES) or low carbon electricity like nuclear energy is at the very heart of the energy transition and European policies as well as many national roadmaps acknowledge its importance. For instance, the RePowerEU plan targets 10 million tons of annual domestic production, plus an additional 10 million tons of annual H_2 imports by 2030 [4]. IRENA's 1.5 °C scenario foresees that H_2 and derivatives will account for up to 12% of final world energy consumption by 2050, but recognises that the cost of production is still a major barrier to the uptake of clean hydrogen [5]. As the cost of electricity represents roughly 70% of the

cost of hydrogen produced by electrolysis [6], it is needed to find low cost electricity. But in addition, electrolysis technologies with improved efficiencies can be very relevant, since they will consume less electricity to produce the same amount of hydrogen. An innovative option uses steam rather than liquid water via the so-called High Temperature Steam Electrolysis (HTSE) process. Based on solid oxide technology (as the Solid Oxide Fuel Cells, SOFCs), it is operated above 700 °C. The use of steam instead of liquid water and the high temperature allows a significant decrease in the electrical energy consumption for the electrolysis reaction [7]. Efficiencies of 84 % $_{el,LHV}$ have been recorded already ten years ago at the scale of a lab system prototype of a few kW [8,9]. More recently, the same level of efficiency has been proven on large scale systems deployed on industrial sites, first at the scale of 120 kW$_{AC}$ [10] and second on a 720 kW$_{AC}$ system [11]. The positive impact of the higher efficiency on the levelized cost of hydrogen (LCOH) as compared to more mature alkaline electrolysis and proton exchange membrane (PEM) electrolysis has been assessed and demonstrated even at the scale of small systems (producing 100 kg/day of hydrogen) [12, 13], with values between 4 and 8 €/kg$_{H2}$ depending on the cost of electricity. For large units producing 500 tons/day, a LCOH around 2.5€/kg$_{H2}$ has been calculated, while for the same plant size alkaline electrolysis leads to a cost of 3 €/kg$_{H2}$ [14]. More recently, for a plant producing 700 tons/day of hydrogen, a LCOH of even 2 $/kg has been reported [15]. The present article focuses on HTSE technology due to its high efficiency and its potential to reach lower LCOH.

However, other levers exist to decrease the LCOH. First of all, decreasing the CAPEX is a key parameter [5], and from that perspective increasing the size of the core technology, the stack, is particularly relevant. While a stack size of a few kW has been considered for many years, the recent trend has been to increase the size of the stack. Traditionally, Sunfire considered relatively small unitary stacks of 30 cells of 128 cm^2 active area [16], but subsequently piled them up [17]. Fuel Cell Energy considers stacks made of 150 cells of 81 cm^2 for mid-size stacks, and 350 cells for full-size stacks assembled in one go [18]. CEA has been developing a stack comprising 75 cells of 200 cm^2 active area [19,20]. Such stacks allow to reach a power of 10–20 kW. As far as the module is concerned, since it is the building block of the electrolysis plant, increasing the number of stacks inside the module, and therefore its power, and considering assembling several modules together while mutualising some Balance of Plant (BoP) components is also an optimum strategy to decrease the CAPEX. While the first system was comprising one single stack [8], modules comprising 4 stacks have been extensively characterized in lab [20–22].

And for the deployment at larger scale in industrial environment, modules with even a larger number of stacks and higher power ratings are considered by the key industrial players. Sunfire is indeed now offering 10 MW units made of 8 × 1.3 MW plug and play hotboxes [23], and Topsoe 20 MW systems made of 48 hotboxes of 480 kW [24]. It is also the case for Bloom Energy and Fuel Cell Energy though they are not disclosing the number of stacks per module. The present article will describe the design of the current generation of Sunfire modules as deployed in the frame of MULTIPLHY project, comprising 60 stacks each, and the assembly of 12 modules and their BoP components, and in particular the Hydrogen Processing Unit (HPU).

Second, the lifetime of the technologies need to be improved [5]. It is a particular working direction for HTSE technology, since most of ageing mechanisms are thermally activated, affecting particularly cells and stacks. The majority of results reported in literature refer to small size single cells, with degradation rates, measured as the evolution of voltage over time while current is kept constant, below 1% or even 0.5%/kh for durations lasting from a few kh to more than 30 kh [25,26]. At stack level, much less results are reported. They mainly concern short stacks, but a few results have been reported at full-stack scale (25–30 cells), with durations up to 9 kh and degradation rates between 0.5 and 2%/kh, the stack being operated galvanostatically as single cells [27,28]. Only recently, another operating strategy has been proposed and applied by

the authors of this article, showing that stacks could be operated at zero hydrogen production loss [29–32].

Last but not least, in order to promote the offtake of electrolysers for the massive production of hydrogen, and in particular for the HTSE technology, which is less mature and less advanced that alkaline and PEM technologies, the installation of MW scale units in representative or even real environment needs to be performed, in order to gain return on experiment and give confidence to early adopters. While alkaline electrolysis units of several MWs are operated for decades, and PEM electrolysers of MW size are now installed for 10 years, both technologies reaching now hundreds and tens of MW respectively, it is not the case for HTSE technology. Only the installation of a 720 kW$_{AC}$ system by Sunfire in a steelmaking plant in Germany could be reported until 2022 [11]. In 2023, Bloom Energy installed a 4 MW in NASA [33].

The present article presents MULTIPLHY pilot unit, with a power of 2.6 MW$_{el,AC,}$ installed in a refinery plant in Rotterdam. It will describe the unit, and present the factory acceptance tests results and the works performed regarding onsite installation. It will also present durability tests performed in lab on stacks of power up to 19 kW, to validate, at such a large scale and for durabilities up to 8 kh, the operation strategy considered to operate the stacks embedded in the modules of the pilot unit.

2. Experimental and procedures

2.1. Description of the HTSE unit and its components

The HTSE unit presents an electrical rated power of 2.4 MW$_{el,AC}$ for a hydrogen production rate of ≥60 kg$_{H2}$/h (≥670 Nm^3/h). Although it is rated at 2.4MW$_{el,AC}$, the modules have in fact been designed for a maximum power of 2.7 MW$_{el,AC}$. Designed and manufactured by Sunfire, it corresponds to a significant upscaling step, MULTIPLHY unit being 17 times bigger than GrInHy unit and 3.6 times bigger than GrInHy2.0 unit.

The modular system is made of 3 blocks of 4 modules. Each module comprises 60 stacks made of 30 cells of 128 cm^2 active area (Fig. 1). Besides the modules, the unit comprises power electronics, placed in a container mutualized with all modules. A fluid interface unit is also part of the unit, to supply inlet flows to the unit (steam, hydrogen, instrument air, nitrogen during transient phases) and to collect products (hydrogen and oxygen). Hydrogen is then sent to the HPU while oxygen is safely vented.

2.2. Description of the HPU unit and its components

Downstream the HTSE unit, a Hydrogen Processing Unit (HPU) is installed. Its aim is to allow the hydrogen produced to meet the quality (at least 3.0) and pressure (30 bar) criteria of the refinery process. This HPU has been designed and assembled by Paul Wurth.

It is constituted of several individual components which are a H$_2$ buffer tank with a capacity of 16.75 m^3, a H$_2$ compressor made of 4 stages, a dryer, an air cooler and a chiller and finally a gas quality online monitoring device (Fig. 2).

The estimated electrical consumption of the HPU is 0.23 MW$_{el,AC}$.

Therefore, the whole unit comprising the HTSE and the HPU parts has a power of 2.6 MW$_{el,AC}$.

2.3. Description of stacks for durability tests and details of the experimental procedure

As part of MULTIPLHY project, durability tests at stack level have been performed in lab, in order to derisk the future operation of the unit and to validate the operation strategy that will be implemented on the pilot unit. Therefore, Sunfire stacks have first been considered for this study, with tests at the scale of a pile of two 30-cell stacks, therefore made of 60 cells of 128 cm^2 active area, for a power around 5 kW –
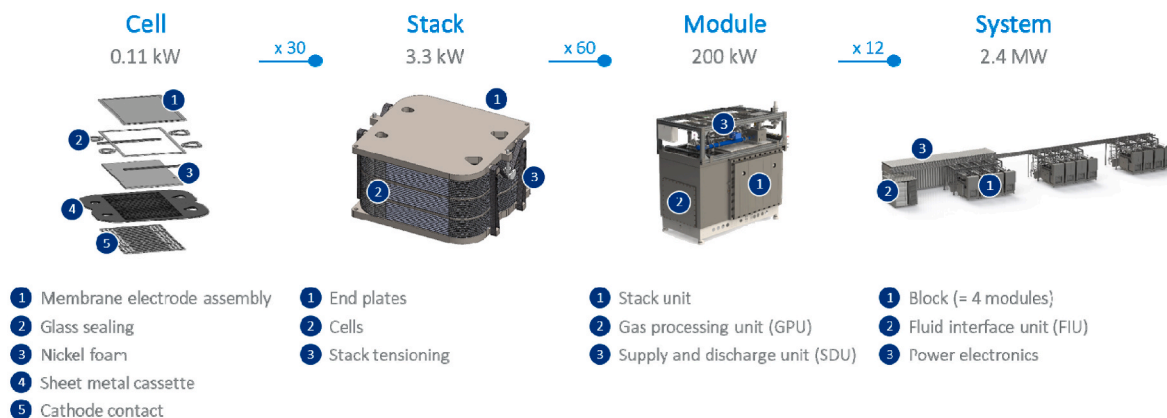
Fig. 1. Description of the 2.4 MW HTSE unit, made of 3 blocks of 4 modules, each module comprising "Gen-2" 60 stacks of 30 cells.
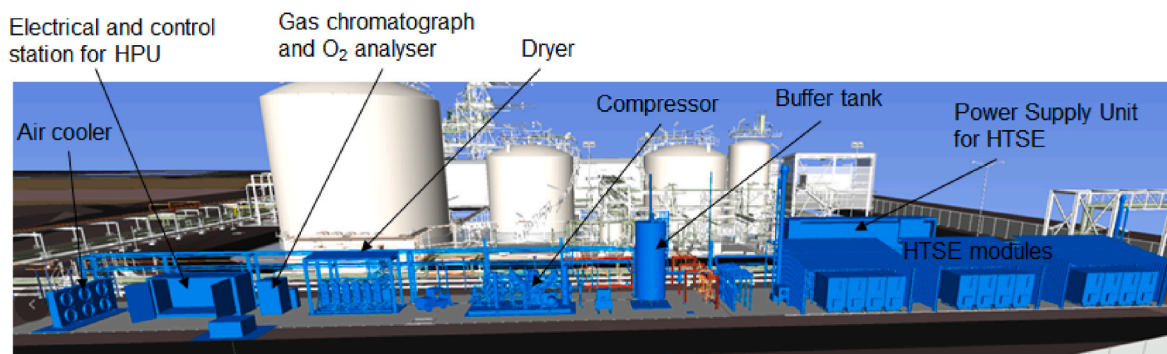

Fig. 2. Description of the HPU unit and of its main components, as installed in NESTE refinery in Rotterdam. The 3 blocks or HTSE modules can be seen on the right.

defined here as "Gen-2" stacks. The Sunfire stacks, made of electrolyte supported cells, is described in detail in Refs. [17,29–31] and is called ESS (electrolyte supported stack) in the following.

In order to validate that the operation strategy developed could be adapted to other types of stacks, a stack made of hydrogen electrode supported cells, so called cathode-supported cells (named here CSS for cathode supported stack) has been tested. Designed by CEA, it comprises 25 cathode-supported cells of 100 cm^2 active area for a power around 3 kW.

For a matter of comparison, a harmonized test protocol has been defined for both types of stacks. It includes the recording of performance maps, several steps at constant current in thermoneutral conditions, as well as load point and thermal cycles. The details are presented in Refs. [29,30]. In this article, we will focus on the durability steps at constant current. It is targeted to operate the stacks in the thermoneutral conditions (that is to say at 1.29V cell at all time), to minimise the thermal gradients and to maximise the efficiency. Therefore, in case of degradation of the performance over time, it means that only a third parameter can be adjusted, which is the stack temperature. Consequently, the operation strategy consists of compensating degradation by a stack temperature evolution over time to keep the average cell voltage approximately equal to 1.29 V and the current density. The initial temperature for each test can be different since it is adjusted to reach the thermoneutral voltage under the given current density and steam conversion. These parameters depend on the initial performance of each stack. The exact values can be seen in the figures of section 4. Nevertheless, the beginning-of-life temperature range was 805–820 °C for ESS, and 675–735 °C for CSS, depending on the stack type/generation but also of the stack history (in some cases stacks have already been characterized before starting durability tests, which can have impacted the performances obtained at the beginning of the durability test).

In a second step of the project, stacks with a higher current density

and/or a higher power have been tested to evaluate the ability of the proposed operation strategy on larger stacks and more severe operating conditions (higher current densities).

On one hand a 25-cell, state-of-the-art "Gen-3" prototype stack from Sunfire was tested at higher current density (-0.79 A/cm^2 compared to -0.5 A/cm for the previous one) - reaching a power of 3.2 kW. On the other hand, two high power CSS have been subsequently manufactured and tested, comprising first 75 and then 78 larger cells with 200 cm^2 active areas. The 75-cell stack was made of 3 substacks of 25 cells [19], and was operated in the same conditions as the 25-cell stack, that is to say a current density of - 0.65 A/cm^2, leading to a power of 12.6 kW. The 78-cell stack, made of a next generation design, was able to operate at - 0.94 A/cm^2, for a power of 18.6 kW [32].

In all cases, stacks were fed with a mixture of H_2O and H_2, with a ratio of 90/10 vol% to prevent oxidation of nickel particles, constituting of the steam electrodes. The inlet flow rates were adjusted to meet a steam conversion of 70% for ESSs and 60% for CSSs. An air flowrate comparable to the steam flowrate was typically used.

3. Results and discussion for pilot unit validation and installation

3.1. Factory acceptance tests on HTSE unit

Before delivery of the HTSE unit onsite, a factory acceptance test has been performed in Sunfire workshop.

The factory acceptance test consists in a series of verifications and validations to check that the unit is in agreement with the specifications and is ready to be shipped for installation at NESTE. The same series of tests has been performed on each of the twelve modules.

It comprised a first cold commissioning step, with basic check of test equipment and interfaces (e.g. opening/closing of valves, …), safety

check (I/O check and Process engineering check for control and measurement loops), and then electrical test (wiring check, grounding test and isolation test). Then each module has been heated up for the hot commissioning step, which included a stack leakage test and an operation phase at rated and maximum load in 90%H_2O/10%H_2 at the fuel electrode and air at the oxygen electrode, during 10 and 12 h respectively. Voltage and temperature were carefully monitored during those stages to check they are situated in the min-max limit set. In addition, a hot idling phase was also performed. At the end of the hot commissioning another leakage test was performed before cooling down. From the hot commissioning phase, parameters such as nominal current density, efficiency, steam conversion, power ramp, hours of stable operation and hydrogen production were checked and should fulfil the specifications defined.

For all of the twelve modules, all quality criteria were passed at the first time, consequently all modules could be shipped to NESTE for onsite installation.

A hydrogen production of 50 Nm³/h has been achieved at part load and of 65.7 Nm³/h at full load. Very homogenous temperatures and voltages were recorded in stacks. A module efficiency above 86 % $_{el,LHV}$ has been recorded at full load, and up to 89 % $_{el,LHV}$ at part load (Fig. 3).

3.2. Factory acceptance tests on HPU unit

Similarly, a factory acceptance test has been performed on each single component of the HPU unit before shipment to NESTE site. The compressor, the dryer and the buffer tank were tested in the manufacturing site to validate the performance specifications.

The compressor test consisted in a mechanical running test with air and the cabinet was visually inspected. The test of the dryer was run with N_2. The chiller was connected to the dryer during the dryer test run. Therefore its operation could be tested as well.

The H_2 tank underwent a visual inspection, a non destructive testing such as Penetrant testing (PT) and Radiographic testing (RT), hardness testing of production welds, and a pressure test, afterwards pickling and passivation was performed. Finally, the analyzers were visually inspected and the valve panel was checked.

As for the HTSE modules, each of the component of the HPU unit passed the quality criteria defined at the first time and could be shipped to NESTE for onsite installation.

3.3. Site preparation and actions performed before startup

Before any installation of the HTSE and HPU units, several preliminary actions have been performed, such as engineering and procurement related activities for the site preparation. Civil and structural works have been first performed. Then piping has been installed, as well as the electrical substation needed to feed MULTIPLHY unit with the 2.6 MW. Instrumentation, automation and control have been defined and designed. Then interface connection has been prepared for the necessary media lines, such as hydrogen, steam, pressurized air and others. Safety approval has been granted before the installation of the units. Fig. 4 presents some steps on the installation of the HTSE and HPU units on site.

After the installation of all components of the HTSE and HPU units, the following tasks have been accomplished before startup.

For the HTSE unit, a cold commissioning (with N_2 and instrument air as media) has been performed, with I/O checks, purging and flushing of lines, calibration of sensors and gas detectors, as well as equipment and group functional testing.

For the HPU unit, a cold commissioning (with N_2 as media) has been performed as well, including I/O check, equipment function tests and group function tests.

These tests were successful and a Pre-Startup Safety Review meeting was organized, leading to the signature of a document allowing the start of the unit.

Then both HTSE and HPU units could be started up. The HTSE was started up (with H_2 and steam as media). Leakage tests were performed. Then all twelve modules were heated up from cold state to hot idling mode. They were subsequently turned into electrolysis operation, by modules in singular and blockwise manner.

In parallel the HPU was started up (with H_2 as media). It underwent a leakage test, and then ran in independent mode, with compression up to 30 barg.
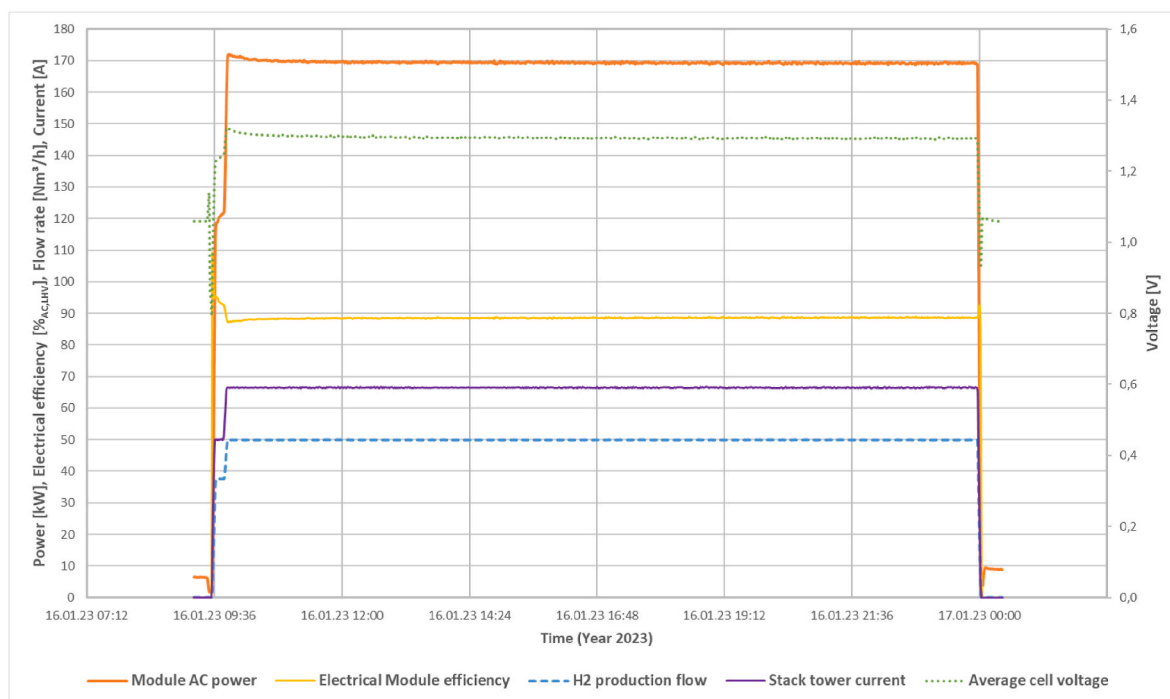

Fig. 3. Power, electrical efficiency, H_2 production flow, current (left axis) and average cell voltage (right axis) recorded on a module within the testing facilities during the hot commissioning step of factory acceptance test, corresponding to the stabilised operation at reference load.
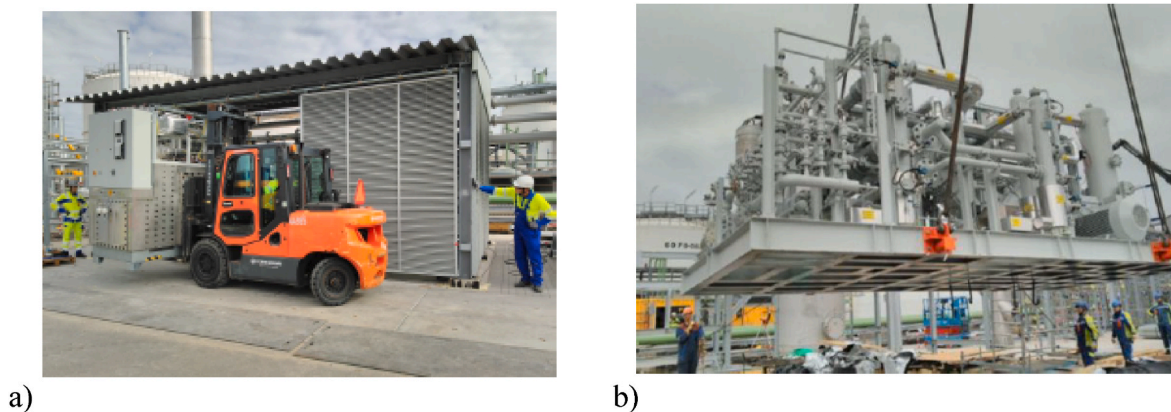
a)   b)

Fig. 4. a) installation of a block of 4 HTSE modules; b) installation of the compressor of the HPU unit.

Finally, the HTSE and the HPU units were coupled, with the hydrogen produced by the HTSE units sent to the HPU.

The achievement of this coupling was a key milestone before the start of the injection of H_2 produced by MULTIPLHY unit in NESTE industrial process.

4. Stack durability tests results

Fig. 5 presents the durability results of Sunfire ESSs, with different numbers of cells, current densities and design generations. Fig. 5a) presents the temperature evolution of a 60-cell "Gen-2" double-stack operated at – 0.5 A/cm^2. Fig. 5b) presents the temperature evolution of a 25-cell "Gen-3" prototype, operated at a current density of – 0.79 A/cm^2. The current densities and operating voltages are kept constant over the entire >8000hrs of operation. On Fig. 5a, the "Gen-2" ESS went through several unplanned events between 3 and 5 kh, caused by failures of the steam supply, and leading to thermal cycles. These events temporarily impacted the stack behavior. Indeed the evolution of temperature following those events did not follow the same slopes as before the event. Those events and induced transient behaviors have not been considered to evaluate the degradation rates discussed hereafter. After normalizing the stack temperature evolution over 8 kh, the overall degradation rate yields +3.9 K kh^{-1}. On Fig. 5b, for almost 8 kh, no noticeable incident occurs to the "Gen-3" prototype stack. The stack temperature averages to an overall rate of only +2 K kh^{-1}. Whereas an increase of the current density would expectedly increase the degradation rate, here, the degradation is lower for the stack operated at higher current density. The design improvements of the "Gen-3" stack are responsible for this preferable result, proving the ability of ESS

technology to operate at higher current density for long lifetime. Notably after approximately 8 kh the stack was shut down for facility maintenance and then restarted (full thermal cycle). Afterwards, the "Gen 3" prototype showed an increased temperature gradient and decreased H_2 output. Regardless of this, the stack continued to operate well (and even partially recover) for another final 1000 h. The source of the observed changes in this prototype has already been identified and addressed by newer design iterations.

Fig. 6 presents the durability results of CSS stacks. Fig. 6a) presents the results of the 25-cell reference stack operated at – 0.65 A/cm^2. Voluntarily excluding the initial stabilization period, linear regressions were performed over a 3100 h segment (between points ❶ and ❷) and another 2100 h segment (between points ❷ and ❸), all points corresponding to stable operation at – 0.65 A/cm^2. As for Fig. 5a), two thermal cycles have been undergone by the stack at 1.5 and 4 kh, which could have impacted its performance. The test was entirely carried out at – 0.65 A/cm^2, except for the step between 3 and 4.2 kh, where temperature increases abruptly to around 750 °C, due to fact that the current density was increased to – 0.85 A/cm^2. This step at higher current density is not considered in the discussion on degradation rate here to allow the comparison between the stacks and the different steps. The rates of temperature increase over time hence calculated were +2.3 K kh^{-1} and +12.4 K kh^{-1}, respectively. Fig. 6b) presents the durability results of the 75-cell stack operated in similar conditions as the 25-cell stack, that is to say a current density of - 0.65 A/cm^2. Its evolution of temperature over time corresponds to +13.5 K kh^{-1}. It is the same order of magnitude as the second step of Fig. 6a), which is rather consistent considering that the 75-cell stack was made of 3 substacks of 25-cells quite similar to the 25-cel stack considered in Fig. 6a), except the cell
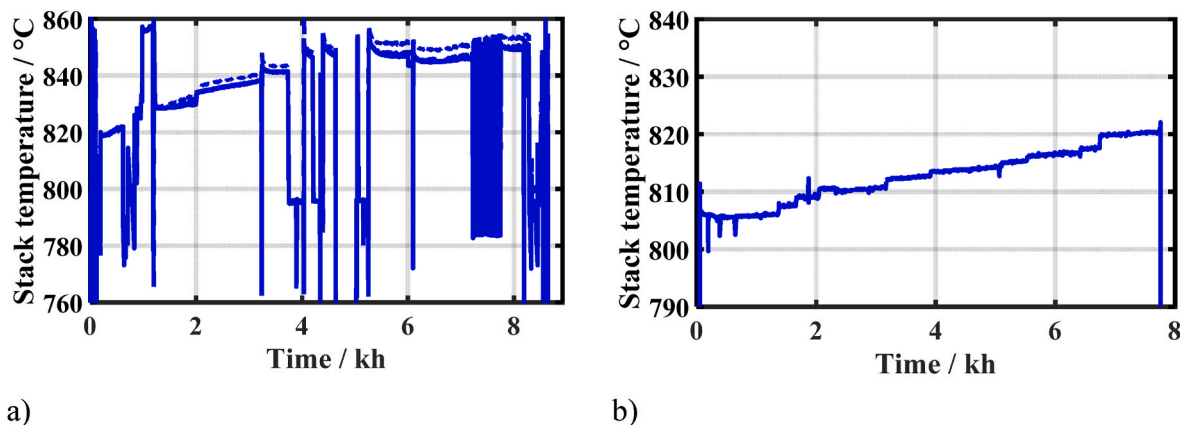

a)   b)

Fig. 5. Temperature evolution curves for Sunfire electrolyte supported stacks operated at thermoneutral conditions. a) "Gen-2" double-stack durability curve, operated at – 0.5 A/cm^2; b) "Gen-3" prototype durability curve, operated at – 0.79 A/cm^2.
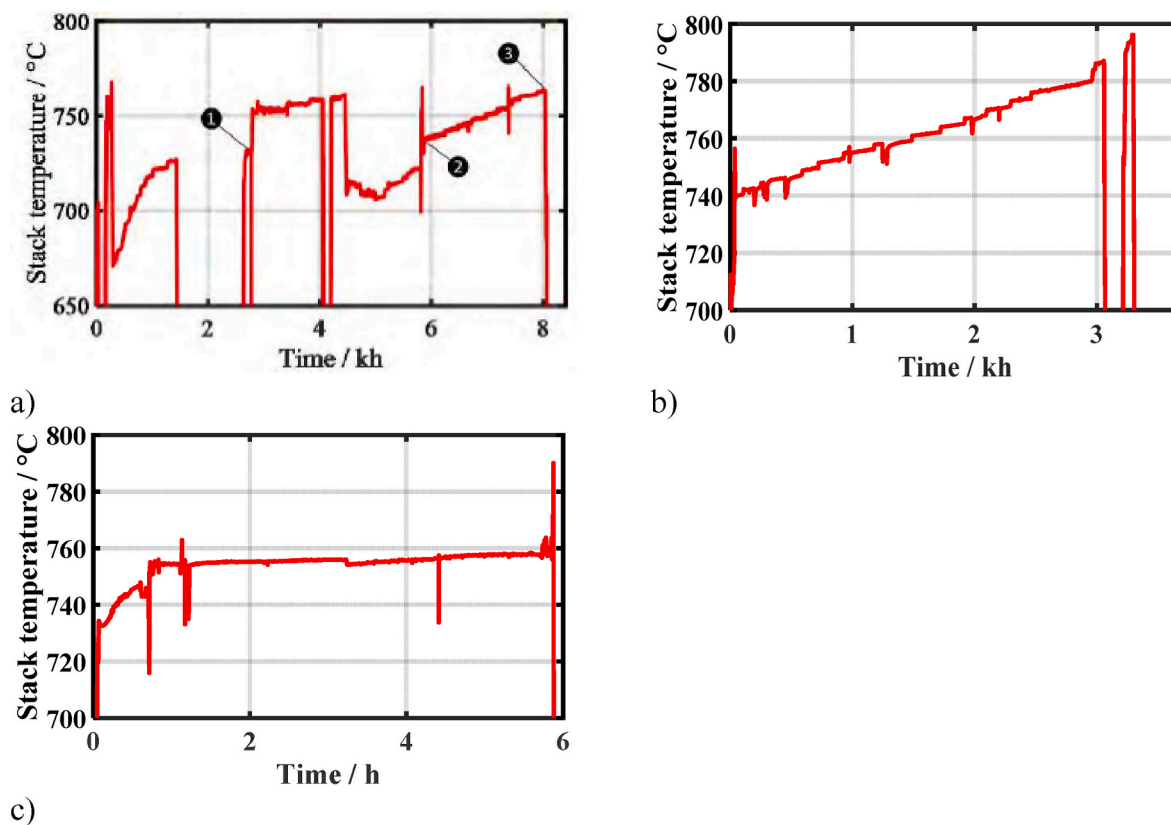
Fig. 6. Temperature evolution curves for CSS stacks operated at thermoneutral conditions. a) durability curve of the 25-cell CSS stack operated at – 0.65 A/cm^2 (except for the step between 3 and 4.2 kh, where temperature increases abruptly to around 750°C, due to fact that the current density was increased to – 0.85 A/cm^2); b) durability curve of the 75-cell stack operated in the same conditions as the 25-cell stack, - 0.65 A/cm^2 ; c) durability curve of the 78-cell CSS stack operated at – 0.94 A/cm.^{2}

area. In addition, this 75-cell stack had already been tested during approximately 1000 h before starting this test, which could explain why the temperature evolution, akin to degradation, behaves linearly and similar to the 2nd step of Fig. 6a) rather than the 1st one. The results presented in Fig. 6c) correspond to a new stack design, assembled in one go, and designed to operate at higher current density, −0.94 A/cm^2 in the present case. Following the transient start lasting approximately 800 h, stack temperature evolved linearly. From 1.0 to 3.2 kh, stack temperature evolved at a rate of +0.92 K kh^{-1}. That rate increased to +1.66 K kh^{-1} from 3.2 to 5.8 kh. Those rates are more than 6 times lower that those recorded on the 2 other stacks considered in Fig. 6a) and b). Given the identical nature of the cells included in both stacks, this improvement is assumed to be attributed to the stack design modifications. These results prove the ability of CSS technology to operate at high current density, above – 0.9 A/cm^2.

5. Conclusion

The 2.6 MW$_{el,AC}$ unit presented in this article comprises a 2.4 MW$_{el,AC}$ high temperature steam electrolyser and a 0.2 MW$_{el,AC}$ hydrogen processing unit. The system is based on Sunfire's "Gen-2" solid oxide technology. The unit is made of 12 modules and is designed to deliver a flow rate of hydrogen of ≥60 kg/h. Each module went through a factory acceptance test and all passed the quality criteria at the first time, and could subsequently be shipped to Rotterdam, to be installed at NESTE renewable products refinery. They were able to demonstrate an electrical efficiency above 86 %$_{el,LHV}$ as expected with this technology. The HPU, responsible to process the hydrogen produced to meet the quality and pressure criteria of the refinery process, has been designed by Paul Wurth and its components also passed the factory acceptance tests with success. On NESTE side, the plot has been prepared in Rotterdam

refinery, to receive the HTSE and HPU units and to provide the requested power line and other utilities. The whole unit has been successfully installed on site and the commissioning performed.

In parallel, long term stack tests were performed at laboratory scale in order to assess the technology performance and durability. Two types of stacks, made of either electrode or electrolyte supported cells, at two scales, a few kW and up to 10–20 kW scale, have been tested over durations up to 8 kh. Thanks to a smart operation strategy adopted for all stack types and sizes and consisting in compensating degradation by a stack temperature evolution over time, it has been shown that they could be operated over those durations without any hydrogen production loss, the current density being kept constant over the whole duration. This operation strategy is the one which will be used for the operation of the pilot.

Thanks to stack design improvement, the latest generations were able to operate at higher current densities, respectively – 0.79 A/cm^2 for the "Gen-3" electrolyte supported stack prototype and – 0.94 A/cm^2 for electrode supported stacks, with a lower degradation rate despite the higher current density. Those results are encouraging for the next generations of the technology, of which the maturity and size of units installed are expected to grow hugely over the next years.

CRediT authorship contribution statement

J. Mougin: Writing – original draft. **J. Aicart:** Data curation. **A. Gajewski:** Data curation. **Anand Kumar Agrawal:** Data curation. **Heidi Bergman:** Data curation. **Pierre Olivier:** Data curation.

Declaration of competing interest

The authors declare that they have no known competing financial

interests or personal relationships that could have appeared to influence the work reported in this paper.

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