Final Report Summary - SOPHIA (Solar integrated pressurized high temperature electrolysis)

Executive Summary:
The project aims at bringing forward the high temperature electrolysis technology by testing it under pressure producing hydrogen, under realistic conditions with heat and steam coming from a solar steam generator, by co-electrolysis for syngas production, combined with developing system concepts for large scale hydrogen and syngas production and an in-depth market analysis.

The potential market for Concentrated Solar Power (CSP) in Southern Europe is estimated between 2 to 4 GW of new installed CSP capacity by 2030 according to the IEA. Combining CSP with a Solid Oxide electrolysis system can produce competitive hydrogen under the conditions of having a grid connection and/or high production capacity. The levelised costs of hydrogen depend strongly on the demand profile and the associated storage capacity needed. Valoration of the by-products oxygen and especially waste
heat will reduce the LCOH₂, as will also a carbon tax will do.

Key targets of the SOPHIA project and expected outcomes are the development of cells (including large scale) and stacks which work under pressurized conditions, meet long durability (< 1% per 1000 h) and high performance (> 1 A/cm²). Additionally the Proof-of-concept for co-electrolysis and syngas production and the manufacture of an optimized stack. These targets have been reached.

Microscopic models, supported by innovative characterization techniques to macroscopic models have been developed. Cell structure improvements as suggested by the modelling results have been successfully implemented by improving the dispersion of two phases in the oxygen electrode functional layer. A resulting gain in performance of 10-15% was observed. By improving the diffusion barrier layer a further performance gain was obtained. Various cells and stacks have been characterised under atmospheric conditions and pressurised conditions. Under atmospheric conditions 7 short-stacks have been tested, one of which for more than 13000 h. Durability of cells and stacks was demonstrated under load-cycling conditions. Degradation rates depend on operation characteristics; they vary from 0%/1000 hr to 7%/1000 hr. It is shown that the Ni coarsening explains around 25% of the degradation rates in electrolysis mode (at 850°C after 1000-2000 h). Besides, the LSCF destabilization is strongly promoted under electrolysis current. Thanks to the modeling approach, a mechanism for LSCF demixing under anodic polarization has been proposed. The numerical tool has been also validated for the pressurized co-electrolysis mode. Cells have also been tested under pressurized conditions both in steam- and co-electrolysis mode. Under pressurize conditions at 10 bar in co-electrolysis mode at the thermo-neutral voltage the performance is better than at atmospheric pressures. A performance better than 1 A/cm² at 1.3 V is observed. A large area stack was successfully demonstrated co-electrolysis. In electrolysis mode the stack was operated more than 1000 hours without degradation. Two 25 cell stacks have been build, one of which was integrated in the system.

A major aim of the project is the proof of principle of a HTSE system at kW size under realistic conditions, i.e. combined with a solar steam generator. This target has also been reached. A HTE prototype system was build including a separate steam generator for stand-alone system testing at 15 Bara. Much attention has been paid to the pressure difference control between anode, cathode and vessel. The solar receiver and steam generator was optimised and able to produce steam at the wanted temperature, pressure and flow. The complete system has been assembled and initially tested with a dummy stack. Pressurised operation at a pressure level of 3 Bara receiving steam from the solar steam generator was successful. The pressure difference between the different compartments could be well maintained during steady state operation as well as during pressurisation. However, the system could not reach its intended operating temperature due to unexpected high heat losses. Nevertheless it was possible to polarise the stack and produce a small amount hydrogen. Following this testing campaign the actual stack was placed in the system for testing in the stand-alone mode. Despite improved insulation it was not possible to reach the operating temperature during heating. However, a small current could be supplied to the stack. The system could well be pressurised up to 15 Bara keeping the pressure difference the anode and cathode well below 50 mbar.

In conclusion the project was successful. A system could be operated with steam coming from solar powered steam generator and at high pressure with a very lower pressure difference between the various stack compartments. Cell and stack performance was improved by adapting the cell microstructure and degradation was reduced. Technology was developed to manufacture large area cells and stacks. A large potential market exists for the SOPHIA technology with production capacities.
Project Context and Objectives:

Context – Europe 2050 plan

In 2010 the European Commission has adopted the Communication “Energy 2020 - A strategy for competitive, sustainable and secure energy”. It includes five headline targets that set out where the EU should be in 2020. One of them relates to climate and energy: Member States have committed themselves to reducing greenhouse gas emissions (GHG) by 20%, increasing the share of renewables in the EU’s energy mix to 20%, and achieving the 20% energy efficiency target increase by 2020.

Together with the White Paper on Transport and the Energy Efficiency Plan, this communication is a key deliverable under the Resource Efficiency Flagship towards the Energy Roadmap 2050. In this roadmap each country is responsible for its own energy choices, which include a mix of sources such as renewable and nuclear energy. The electricity production in Europe between 2010 and 2030 will grow from 3335 TWh to 3706 TWh, and the part from renewable energy sources should increase from 21% to 46%.

To ensure the evolution of total electricity production for the next few years, the total installed capacity of RE is forecasted to increase from 243 GW (in 2010) to 476 GW in 2020 and 613 GW in 2030.

There are eight main RE technologies in the European portfolio for electricity production: wind on- and offshore, large and small hydro, biomass, geothermal, photovoltaic (PV) and Concentrated Solar Power (CSP). Wind and solar power are intermittent sources of electricity with limited predictability.

In fact, generating power from renewable energy sources is subject to extreme fluctuations and cannot be controlled since the period during which power is generated (when the wind is blowing or the sun is shining) does not always coincide with power consumption. Today already, not all green energy generated can be fed into the electric grid at the same time.

Therefore the large-scale deployment of renewable energy requires an energy storage solution in large quantities. This storage can also play a role in the regulation (e.g. frequency) of the electrical grid.

High Temperature Electrolysis a part of the solution for reaching Europe’s 2020 and 2050 goals

High Temperature Electrolysis (HTE), which produces hydrogen from water and electricity or syngas from water, CO₂ and electricity, is one of the solutions that will help Europe attain its 2050 targets in a “power to gas” vision.

“Power to gas” consists in taking profit of the intermittent overproduction of renewable electricity (RE) at low cost and optionally CO₂ from industrial facilities to produce high value products such as hydrogen, syngas, synthetic methane (or methanol, or DME) and therefore store energy.

Energy conversion technologies like CSP and nuclear offer the advantage to provide power and heat at the same site. The coupling of a heat source with HTE allows the latter to reach very high efficiency levels, which is a major differentiator to other electrolysis technologies.

Industrial sectors that produce a lot of waste heat generally also emit CO₂. Examples are the steel industry and the cement industry. Such sectors are favorable for drawing co-electrolysis scenarios where CO₂ is valorized into syngas that can then be transformed in other value-added products.

These sources of heat and CO₂ are widely available and spread across Europe and very often they are very close to power sources.

For instance in France there are two main zones where the electric grid will not be able to absorb all electricity produced issued from intermittent renewable energy like wind and solar power. These two
zones are the northwest and the south. These renewable power source regions also include many heat and CO2 sources. In other countries like Germany the situation is similar: the electricity grid is currently not, and will not be in the near- to mid-term future, able to transmit the surplus of renewable energy from the north where it is available to the south where it is needed.

There is a large number of applications in a “power to gas” vision, among which the following examples:

• Fuel cell and hydrogen technologies are part of the European Strategic Energy Technology Plan. Hydrogen produced by electrolysis and used for transport applications contributes to lowering greenhouse gas emissions and the dependence of Europe on fossil mobility fuel. Several car manufacturers have announced the commercialization of H2 vehicles embracing Europe’s vision.

• Hythane makes storage and transport of hydrogen easy: hydrogen can be introduced in the existing natural gas network (making Hythane) for domestic applications, lowering GHG emissions of those.

• Energy storage favors the deployment of renewable energy by introducing flexibility into the electrical network and helping offer meet demand. Storage also allows for a high electrical network efficiency by ensuring that all energy produced is consumed.

• Electrolysis of water and CO2 (co-electrolysis) produces syngas that can be further transformed in many end products such as synthetic methane, methanol and others. These products can be used as a fuel like methane or by industries like the chemical industry. Co-electrolysis coupled with renewable or nuclear power is not only a way to produce fossil fuel free products; it can also consume and valorize CO2 emitted by industries such as steel, cement, and domestic waste incineration, which are numerous and spread over Europe.

Concept – taking HTE further towards large scale deployment

The project SOPHIA is built for bridging the gap between highly encouraging proofs of concepts (POC) (e.g. proven in the FP7 RelHy project and the FCH JU ADEL project) and the requirement of the development of reliable HTE pressurized systems of high durability and performance, for hydrogen or syngas production necessary for the future EU targets.

More efficient HTE systems will require high temperature pressurized operation in order to avoid additional hydrogen pressurization just after production. First published experiments on pressurized cell testing show promising results with an increase in the kinetics that compensates the less favorable thermodynamic conditions. In addition, the most performing generation of cells and stacks is to be further developed to pressurized conditions and up-scaled.

There are three main development lines (cell, stack, and system) and one concepts line in the SOPHIA project.

Transversal to the three development lines are testing and modeling activities. Micro-scale to macro-scale models are developed aiming at understanding the underlying mechanisms for pressurized electrolysis and co-electrolysis. Based on the results of previous activities in the EU funded project Hi2H2 and the FCH JU project ADEL system concepts are elaborated to couple the HTSE system to a renewable energy source.

The key targets of the two first development lines are to develop cells (including large scale) and stacks which work under pressurized conditions, meet long durability (< 1% per 1000 h) and high performance (> 1 A/cm^2) targets. These two lines are:
1) Large scale and optimized cells for SOE

Cells will be developed with tailored electrode microstructure and compositions, specially designed to work under electrolysis and co-electrolysis conditions. The improvement of the microstructure is based on the outcome of model calculations for the micro-structural optimizations, aiming to improve the electrode efficiency and second to reduce cell degradation and microstructure analysis before and after operation. In addition, the design of the supporting electrode will be optimized to reach the mechanical stability required for integration in the pressurized stack. The cell architecture consolidated by the validation step will be used for the production of 100 cm² active cells for the pressurized stack and 200 cm² or more for the large area atmospheric stack.

2) Optimized stack

Solid oxide steam- and co-electrolysis at increased pressure will be studied to achieve the necessary understanding and to select high performance and durable cells and stacks for further up-scaling in terms of power and area. The strategy in this project is therefore to operate optimized stacks (with regard to cell, stack components and control strategy) both in steam electrolysis mode to produce H2, which can be used directly or which can be used as one of the reactants in synfuel synthesis, and in co-electrolysis mode of steam and CO2 to produce synthesis gas which can be used directly as the reactant in synfuel synthesis. Based on the experimental performance and durability results on cells and short-stacks, combined with the modeling, two optimized stacks will be designed and prototyped.

Tests will be performed on cells and short-stacks under pressure in electrolysis and co-electrolysis as a proof-of-concept for co-electrolysis under pressure.

A large area stack (cells of at least 200 cm²) will be manufactured and tested in electrolysis and co-electrolysis mode to prove the up-scaling capability of the technology.

A 3 kWe-stack operating under pressure up to 15 bars will be prototyped and tested in electrolysis mode since an increase of the cell performance and a better integration of the SOEC module with up- and downstream processes are expected; it will be coupled to a concentrated solar energy source for on-site proof of concept.

The third development line aims at proof of principle of a HTSE system at kW size under realistic conditions (high humidity, high temperature, pressurized, fluctuating production).

The concept proposes the development of an optimized system coupled with concentrated solar power as a source of heat and power, to be deployed on-site. An optimized HTSE set-up will be identified and designed. It will be realized and equipped with all necessary Balance-Of-Plant components needed to carry out a full proof of converting water and renewable energy into hydrogen. After qualifying such electrolyser system first under laboratory conditions with defined and controlled boundary condition, especially heat and electricity input, in a last phase of the project this system will be operated on-sun, powered by solar energy from a solar furnace applying real boundary conditions.

A comprehensive “concepts” line aims at developing concepts of HTE for use in connection with renewable energy production (wind, solar) and nuclear power. Several case studies will be developed using results derived within SOPHIA.

For each case study, a completed and integrated process chain will be defined in function of main SOPHIA results obtained during market analysis that will be focus on the energetic context, CO2 resources and specifications (and its characterization) and add-value products market evolution and its specifications (methane, hydrogen or liquid fuels). These results will allow us to define, for each case
study:
• the plant location (after a local context analysis),
• the plant size,
• the add-value product to valorize, etc.
• the main technical and economic hypothesis (period of electricity cost, etc.).

A particular focus will be done in this task on the integration of the developed concepts with the industrial 
CO2 emitters, oxygen consumers and potential customer’s plants.

Project Results:
The project is divided into 6 scientific and technology development work packages, one for dissemination and 
exploitation and one for project management. The six S&T work packages will be addressed in the 
following including the market analysis done in the dissemination and exploitation work package.

WP2: SOE concept Development
The main objectives of WP2 are:
• Definition of the industrial specifications of High Temperature Electrolysis and Co-Electrolysis systems 
for the production of hydrogen and syngas that will be converted into carbon containing fuels (e.g. 
synthetic methane and liquid fuels).
• Definition of a System Requirements Document (SRD) with technical end-user requirements for the SOE 
module and overall system and specific technical requirements of the prototyping unit will be produced.
• Identification of the different scenarios for a complete process chain for the technological concept 
development and its end-products valorisation.
• Techno-economic analysis on the different case studies identified for concepts industrialization 
development.
• Environnemental analysis son more relevant case studies

WT2.1 Industrial specifications for a prototype of a pressurized electrolyser system coupled with 
concentrated solar
Within the project, an improved high-temperature electrolysis and co-electrolysis technology for coupling 
to solar power sources is developed. A 3 kWe prototype electrolyser unit will be designed, constructed and 
operated and combined with a Concentrated Solar Power system receiving its energy from the solar 
simulator at DLR. The prototype unit will contain newly developed stack technology with balance of plant, 
and operate under elevated pressure (up to 15 bar). The end-user requirements for this technology as well 
as the system requirements for the prototype system are developed.
Prior to developing the requirements a proper description of the SOEC technology and solar thermal 
technologies, has been made and the important characteristic system parameters, such as operation 
mode, steam conversion and the use of a sweep gas have been discussed. Especially a description of the 
prototype equipment to be applied and further developed within the project is made (both of the SOEC 
stack and of the solar simulator).

The 3kWe HTE prototype is too small to be tested in a solar tower unit. Therefore, it is coupled with a solar 
tubular receiver developed at DLR and tested in the DLR’s solar simulator. In a receiver, the radiation 
passes only from one side through an aperture and irradiates tubes through which a heat transfer medium 
is led. This minimizes heat loss even at high temperatures. A solar tube-type receiver with a cylindrical 
configuration has been thus developed in order to provide the electrolyser with superheated steam.
The stack needs to be kept hot all the time, also over the night which can be an integration issue for the coupling of the HTE with the solar energy. This issue can be solved by addition of some sufficient heat storage capacity or the eventual addition of electrical heaters. Concerning the prototype of 3kWe HTE which will be tested in the solar simulator, an electrical heater may be added in order to keep the stack hot all the time.

Requirements depend on the application of the hydrogen: three applications are considered, i.e. injection into the natural gas grid, transportation and production of chemicals by methanation of CO2. Purity requirements are somewhat comparable, setting limits mainly to H2O levels in the product. No major other contaminants are expected from the SOEC unit. The required product pressures differ but are all above atmospheric. For large scale applications it will be required to deliver the hydrogen continuously (i.e. 24/7); for transportation discontinuous production might be feasible if storage capacity is high enough.

The prototype will be a 3 kWe electrolyser; the actual power may vary between 3.0-4.8 kWe, depending on amongst others the operation temperature. For a 25 cell stack operating at a cell voltage of 1.30 V the stack voltage will be 32.5 V (DC) (in autothermal mode) at a stack current of 100-150 A, yielding a H2 production of 1.0 Nm3/h.

Electricity will be taken from the grid. The system will include a power conditioner to deliver DC power at the required voltage. Dynamic power variations may be invoked on purpose as part of the test program, to simulate the dynamic behaviour of large-scale CSP systems.

Product storage is not necessary; the products (H2 and O2) shall be vented into the atmosphere. Sample lines shall be included to enable chemical analysis. Flow meters shall be installed to determine production capacity at given power input.

Due to the safety regulations at the testing location (DLR), operation shall cease at night, meaning that the system shall be put in a stand-by mode. Provisions have to be included to keep the system hot, such as electric heaters for continued generation of hot steam that is to be fed to the system. Most likely the pressure has to be released for stand-by operation, due to the safety regulations at DLR.

The system shall be controlled automatically by means of a microprocessor, but with a proper human-machine interface. Start-up and shut-down procedures aim at slow heating/cooling avoiding temperature shocks. The system shall include all required mechanical safety devices to avoid over-pressurization and unwanted back-flow of hazardous gases. Moreover, an emergency shut-down procedure shall be devised to cover amongst others any of the following situations: Over-pressure, Loss of pressure, Over-temperature, and Fire.

**WT2.2 Concepts development for electrolysis and co-electrolysis coupled with power (renewable and other energies), heat and CO2 sources**

This task is dedicated to the definition of case studies that will represent potential strategies for the industrial concept development. A particular focus in this task lies on the integration of the developed concepts with the industrial CO2 emitters, oxygen consumers and potential customer’s plants.

Given the preliminary results of the market analysis, three countries present important potential for the SOPHIA concept and those countries are France, Spain and Italy. They actually have interesting average DNI (Direct Normal Irradiation) up to 1800 kWh/m2 which allows an efficient operation of the CSP
(Concentrated Solar Power) process brick plus the energetic context is favourable to added value products such as hydrogen, methane, methanol or liquid fuels (as gasoline).

The 5 case studies are listed and described below:

• Case study n°1 – FRANCE / hydrogen production for mobility market (SOE pathway): the best location for France in terms of DNI is in the south east of France (PACA region). The production capacity is supposed to be 400 kgH2/day which roughly corresponds to HRS (Hydrogen Refuelling Stations) capacities at the beginning of HRS development. Hydrogen purity will need to be 99.999% and should be delivered at 350 bar. Currently, few HRS are in operation but a massive deployment is expected with 600 stations by 2030. Thus, production capacities of 4 000 and 20 000 kgH2/day will also be analysed. Operation time is preliminary fixed at 3000 h/year which corresponds to 8h a day sunlight during 365 days. In case SOPHIA plant need to increase its production (during the night for example) electricity market was investigated and we found that electricity average annual market price was 34.6€/MWh in 2014 For France and prospective prices were estimated by 2030. Finally, given the renewable agenda in France, many hours of electricity surplus are expected by 2030 which might be interesting in case to increase SOPHIA plants operating time with low cost operating expenses.

• Case n°2: Italy – methane production for mobility market (SOE pathway): Given the DNI, the Lazio region was selected amongst numerous high DNI regions because here we also can find many industrial CO2 emitters. Given the NGV market in Italy which is the most important in Europe a 4 000 and 20 000 kgH2/day capacities plants are considered operating 3000 h/year. Produced methane will need to respect specifications of the future European Norm (EN16723) and will need to be delivered at 200 or 250 bars. The industrial considered for the CO2 source is a glass manufacturer (~75ktCO2/year and [CO2]~25-35%vol which can be a potential consumer of O2 co-produced within the SOPHIA plant.

• Case n°3: France – methane production for injection in the natural gas grid (SOEC pathway): the selected location is the same than case 1 for the same DNI reason and also because of a high concentration of industrials CO2 emitters. The plant capacities are 4 000 and 20 000 kgH2/day and the produced methane will need to respect future European norm (EN16723) and should be delivered between 4-16 bars for distribution grid and between 40-70 bars for transport grid. The industrial considered as a CO2 provider is a steel manufacturer (~7990 ktCO2/year and [CO2]~30-40%vol.) which can be an important O2 consumer.

• Case n°4.a: Spain – methanol or gasoline production (SOEC pathway): The selected location is Andalucia. The CO2 provider is considered to be a petrochemical company (~1520 ktCO2/year and [CO2]~10-15%vol.). This company was chosen because it can be an O2 consumer and also a client for the final product of SOPHIA plant.

• Case n°4.b: Spain – hydrogen production for mobility (SOE pathway): this case study is identical to case 1 except for the location (and so economic conditions). It will permit to assess to the sensibility of an economic context on the final price of a product done by the same plant.

WT2.3 Industrial specifications for large scale electrolysis and co-electrolysis systems derived from the developed concepts

Based on the selected sizes for the industrial application some preliminary specifications have been made. It is assumed that for an industrial application a continuous operation is needed, which implies that there will be a need for a heat storage and power source during the night. Typical values of the system parameters for the transportation application and the industrial use are 400 to 4000 kg/d.
The quality requirements of the produced hydrogen will depend on the application; H2 into NG grid, H2 for transportation, H2 for methanation. It is expected that hydrogen produced from steam electrolysis has a high quality with as main impurity steam; about 40% in the hot hydrogen (depends on steam conversion), and below 1% after cooling/condensing (15 bar, 45 °C). For end applications the dew-point requirements usually are very strict (~10 ppm), hence an additional dryer would be needed.

WT2.4 Development of flow sheeting derived from industrial specifications

Flow sheets have been developed for industrial application for scaling up solar-driven SOE plants and Power-to-Gas/Liquid plants. Being given the importance of a good storage for the fully solar-driven HTE system, it has been focused in this task on the coupling of pressurized high temperature (co-) electrolyser with the molten salt solar tower technology due to its higher storage capacity and on the optimized corresponding flow sheets.

First, a simple (co-) electrolyzer modelling approach was implemented. Then, the rigorous (co-) electrolyser modelling developed by the consortium has been fully integrated to the global process including, among other functionalities, the molten salt loop. Coupling with the molten-salt solar tower technology, both the hydrogen production by the electrolysis of steam and syngas production by the co-electrolysis of steam and CO2 have been analyzed for HTE system. The processes were analyzed from a thermodynamic point of view by calculating the overall process efficiency defined as the higher heating value (HHV) flow of the product in relation to the thermal energy input. The processes have been designed independently of the location with the aim of minimizing the required thermal energy input, i.e. maximising the thermal-to-fuel efficiency. For each case study, and in order to enhance the economics of the process, thermal energy storage has been considered in order to increase the electrolysers operating time beyond the hours of sunlight.

In a second step the rigorous electrolyzer model developed by VTT has been implemented in the global process configuration proposed by DLR for the production of renewable hydrogen from CSP energy. Simulations have been performed in order to assess the technical feasibility of a process producing 4 000 and 20 000 kg/d of hydrogen.

A further conversion of syngas or a mixture of H2 and CO2 to carbon fuels or chemicals has been studied for large-scale application of (co-) electrolysis technology. Particularly, methane production was highlighted and studied due to existing infrastructures for massive storage and transportation. The production of liquid-form chemicals or carbon fuels, e.g. methanol and gasoline, was also investigated due to the applicability of large-scale storage and available technologies. Rigorous models concerning the process chains leading to methane and liquid fuels production from were also implemented in overall process flowsheeting that will be used in techno-economic assessment task.

WT2.5 Techno-economic analysis and LCA

Regarding the 5 cases studies proposed and the flowsheeting development, the techno-economic assessments have been performed for all the cases for three production capacities: 400, 4 000 and 20 000 kgH2/day. This economic evaluation is completed by a life cycle analysis, to see to which extent these solutions for solar energies valorization are relevant from an environmental point of view. Results obtained from the techno-economic analysis show that the local solar irradiation is one of the most significant points for location selection. The three locations identified are all with quite high average irradiation; however, the irradiation in Spain is much more even and kept at a high level all over the year,
which potentially reduces the oversizing of CSP and the size of product storage, thus further reducing the product cost.

Concerning the H2 production, it was observed that the techno-economic optimal configuration is when the chemical process part of SOPHIA plant works with a baseload, the intermittency of the solar power generation being smoothed by the CSP process part. This leads to a high operating time ratio of the plant (around 23 h/d in average over the year). However, this configuration does not match to the best energetic configuration, as the CSP part is oversized, meaning that additional high temperature heat can be valorized. Within these conditions of base load working point for the H2 generation process, the use of PV panels + batteries does not represent any advantages, as the storage (batteries) is very expensive compared to the thermal storage of CSP. The H2 production for mobility application is identified to be competitive when the plant capacity is sufficiently important (beyond 4 000 kgH2/d), and when the grid access is authorized, to avoid important CSP oversizing. These results are strongly linked with the H2 consumption profile, assumed constant over each hour of the year.

Concerning methane, methanol and gasoline production, the same trends have been observed in terms of impact of solar irradiation on the overall production, the production capacity and the integration of complementary electricity sourcing as grid and eventually PV. Due to the low annual capacity factor of PV compared with CSP, which allows cheap and massive thermal storage, PV is generally not preferred to combine with SOE for energy storage from a cost-effectiveness viewpoint, particularly when its annual capacity factor is below 20 %. However, when above 20 %, PV seems to become competitive with grid power. To minimize the levelized product cost, the CSP tends to operate as long as over 22 hours for most plants, with the aid of cost-effective and efficient molten-salt thermal storage. When increasing the annual power share of CSP, basically the whole system is oversized allowing to produce more products during summer time, thus leading to a larger seasonal storage and a higher product cost. For each product, the levelized product cost is mainly dominated by the CAPEX (75 % - 85 %), with the remaining OPEX mostly contributed by equipment maintenance and grid power use (if it is the case). At least 60 % CAPEX comes from CSP (depending on the case study), followed by SOE (~10 – 15 %) and heat exchanger network (~10 %).

Concerning the life cycle analysis, best results show that SOPHIA process produces a hydrogen with much lower GWP impact: 1,06-2,31 kgCO2eq/kgH2 (4 000kg/d configuration) vs. 11,9 kgCO2eq/kgH2 reported in literature for the SMR. Best results are obtained with the CSP+grid energy sourcing, as the CSP-only case appears to suffer from a reduced efficiency probably related to the excess heat produced and not valorised. In addition, this case requires massive H2 storage with potential significant GWP impacts from the preliminary estimate performed. Even with such storage however, SOPHIA results remain significantly better than hydrogen production by SMR.

Sensitivity analysis on the electricity mix showed the importance to leverage low-carbon electricity when sourcing energy from the grid. With the use of a Spanish average mix to complement CSP, with a carbon content of 466 gCO2eq/kWh, hydrogen from SOPHIA increases to 6,6 kgCO2eq/kgH2. French electricity mix is much less impacting, due to its nuclear component, leading to only 2,0 kgCO2eq/kgH2.

WP3: Electrochemical Modeling & Validation
1. Introduction
The main objectives of WP3 are: (1) to understand the SOE electrochemical mechanisms for pressurized steam electrolysis and co-electrolysis, (2) to propose some recommendations of optimized electrode microstructures regarding the steam electrolysis and pressurized co-electrolysis, (3) to estimation of the
risk of degradation (ageing in operation) and (4) to specify some optimized operating conditions. In order to reach all these targets, a methodology based on an integrated experimental and modelling approach was proposed. The main results obtained in the frame of WP3 are summarized in this section.

2. Electrode microstructural characterizations and modeling

2.1 Synchrotron X-ray holotomography

The synchrotron X-ray nano-holotomography method has been adapted to the highly absorbent ceramic materials of SOCs. This technique allows obtaining relevant reconstructions with a high field of view and a high spatial resolution. Both of these features of the 3D volumes are crucial to describe accurately the SOC microstructure. In this frame, a new sample preparation using a specific Xe+ Plasma Focused Ion Beam (PFIB) equipment has been developed. Thanks to the high precision and throughput of the equipment, specimens have been prepared with a section as large as 50μm. The sample geometry presents a controlled axisymmetric shape which is especially well adapted for tomographic measurements. The experiments have been performed at the new Nano-Imaging beamline ID16A-NI at the European Synchrotron Radiation Facility (ESRF). This new beamline has been designed to obtain three-dimensional reconstructions from large field of view while maintaining a high spatial resolution. By improving the process of data acquisition, electrode reconstructions have been obtained with volumes of $\approx 51 \times 252 \times \pi \text{ μm}^3$. The spatial resolution was increased to approx. 50 nm in order to accurately describe the electrodes fine microstructure.

2.2 Microstructural computations

Standard measurements – Electrodes reconstructions have been obtained for the pristine cells and after long-term operations. All the electrode microstructure features, such as the mean phase diameters, the phase specific surface areas or the triple phase boundary length (TPBls) have been quantified on the reconstructed 3D volumes and used in the multiscale models (cf. sections 3).

Notion of accessible TPBs – As the previous standard 3D measurements partially inform on how local geometry and network topology causes variability in TPB accessibility, a new concept of ‘accessible’ TPBs has been proposed. This concept has been defined as the measure of the effective conductivity of the ionic, electronic and pore transport path networks between each TPB and the electrolyte, current collection and gas channels, respectively. This notion has been applied to the Ni-YSZ cermet. It has shown that a systematic error is introduced in the simulations if assuming the microstructure is depicted as a continuous medium represented by its effective properties. Therefore, in complementarity with the standard measurements, the concept of accessible has been also used to characterize the electrode microstructure evolution after cell operation.

3. Reactive pathway and role of microstructure for the O2 electrode operated in electrolysis mode

3.1 Electrode micro-scale modeling and validation

Since the operating mechanisms of LSCF and LSCF-CGO electrodes were not still understood in electrolysis mode, a special attention has been paid to propose a modelling framework that includes a relevant reactive pathway explaining the performances of such materials in anodic polarization. The model takes into account the microstructure properties of the electrode phases as well as the most likely processes occurring therein. The reactive pathway has been divided in a sequence of elementary steps with a first oxido-reduction at the gas/MIEC surface (bulk path), and a second one in parallel at the electrode TPBs (surface path). The model has been validated on experimental data recorded at different
temperatures on a LSCF electrode tested in a symmetrical cell configuration. Thanks to the 3D electrode reconstruction, all the microstructural properties required for the simulations were determined, and thus, the number of unknown parameters for the model was considerably reduced. In these conditions, a good agreement was found between the experimental data and the simulations with fitted parameters lying in the range of their expected values.

3.2 LSCF and LSCF-CGO reactive pathway in electrolysis mode
Once the model validated for the LSCF single-phase electrode, simulation results have highlighted that the reactive pathway is governed by the charge transfer at the gas/MIEC surface (bulk path) in cathodic polarization. The mechanism is found to remain unchanged at low anodic polarization, whereas oxidation at TPBs (surface path) becomes the predominant reactive pathway at higher polarization. For the LSCF-CGO composite, simulations have shown that the reactive mechanism is governed by the charge transfer at TPBs whatever the electrode polarization.

3.3 Microstructural optimization
The effect of electrode microstructure on the cell response has been studied by sensitivity analysis for both the single-solid phase LSCF and LSCF-CGO composite electrodes. The numerical analysis has revealed that some of the morphological properties can have a huge impact on the electrode performances. For the single solid phase electrode, the electrode specific surface area (between MIEC and gas) is found to have a strong impact on the performances in both cathodic and anodic polarizations. The ratio of the volume fraction to the tortuosity factor for the MIEC phase has a non-negligible impact under cathodic polarization (while its effect is rather negligible under anodic polarization). Moreover, the LSCF-CGO composite presents a much higher performance compared to the LSCF single phase electrode, especially in anodic polarization. In that condition, an electrode made of a LSCF-CGO composite should be particularly relevant for the electrolysis mode. These results have been used as guidelines for microstructural optimization in the project.

4. SRU modelling: impact of pressure in steam and co-electrolysis
4.1 Model and validation
The micro-scale electrode models have been connected with an available macro-scale module allowing the computation of the global SRU response. For the cell of the project, it has been checked that the global numerical tool is able to accurately simulate the i-V curves at Pt=1 atm under different operating conditions. The model has then been adapted to take into account the effect pressure in steam and co-electrolysis. It has been assumed that pressure effect on the cell response is mainly due to the impact of Pt on the OCV and on the concentration overpotentials (due to the mass transfer trough the electrodes). The updated model has been validated at 10 bars in both modes. It has been found that the model is able to predict accurately the polarization curves as well as the syngas compositions at the cell outlet.

4.2 Impact of pressure on cell operating mechanisms
Once validated, the model has been used to analyze the operating mechanisms under pressurized conditions. It has been shown that the improvement of the cell limiting current density under pressure is ascribed to a decrease of the gas diffusion resistance across the electrodes. At the same time, it has been found that the convective flow through the porous electrodes is negligible whatever the operating pressure. In co-electrolysis, the simulations have shown that CH4 production under pressure is favored in an
electrode volume located at the cell outlet close to the electrolyte interface, where H₂O and CO₂ are mainly electrochemically converted into H₂ and CO. These analysis has clearly highlighted the close interaction between the electrochemical and chemical reactions for the internal production of CH₄ within the cermet.

4.3 Optimization of the operating conditions under pressure
Considering the cell of the project, simulations have been carried out to identify the best operating conditions under pressure in steam and co-electrolysis modes. In steam electrolysis, it has been found that the hydrogen production is higher under pressure at 1.3 Volts, even until 30 bars. In co-electrolysis, relevant operating conditions have been identified at 10 bars: at 700°C and 1.3 V, an inlet cathodic flux of around 10 Nml.min⁻¹.cm⁻² yields to a slight exothermic mode resulting in a reasonable faradaic conversion rate of 60% (for a current density of -0.80 A.cm⁻²). With this operating conditions, the CH₄ content in the syngas is restricted to around 5% on the dry outlet gases with a ratio of H₂/CO=2.25. The same analysis has been also conducted at 30 bar. In this case, the cartographies have shown that formation of CH₄ in the co-electrolyser remains limited even at 700°C and 30 bar.

5. Cell degradation in electrolysis versus fuel cell modes
A set of long-term tests have been carried out in electrolysis mode in the frame of the project. In synergy with Endurance project (FCH-JU-2013-1, Grant agreement n°621207), the degradation tests have been complemented with durability experiments performed in fuel cell mode. In similar operating conditions, the degradation rates were found to be higher in electrolysis than in fuel cell operation.

5.1 Study of Ni coarsening upon ageing
Reconstructions of the Ni-YSZ cermet have been obtained before and after durability tests. Electrode microstructure evolutions were found to be similar after 1000h operation in fuel cell or electrolysis conditions. More specifically, the evolution of the accessible TPB measured during operation shows that SOFC mode for 4700 h does not alter how the electrode microstructure is utilized. The degradation in SOEC mode for 10700 h is in contrast noticeable, but the shape of the distribution is preserved. Nonetheless, a clear increase of the Ni particle diameter over the time has been detected: the higher the operating temperature, the higher the Ni agglomeration. These results allow relating the Ni phase coarsening upon operation to a sintering process thermally activated. Finally, it was computed with the model that the significant loss of electroactive sites due to the Ni coarsening is liable to explain between 20 to 30% of the cell performances degradation.

5.2 LSCF destabilization and Sr reactivity with the electrolyte
The active layer of the O₂ electrode made in LSCF and CGO is liable to undergo material decompositions and reactivity with the YSZ electrolyte. As LSCF reactivity with the electrolyte reflects the overall materials destabilization, a special attention has been paid to characterize the secondary phases after ageing. For this purpose, (i) Scanning Electron Microscopy (SEM), (ii) Transmission Electron Microscopy (TEM) coupled with Energy Dispersive X-ray (EDX) analyses, and (iii) a specifically adapted method based on synchrotron X-ray µfluorescence and µdiffraction tomography have been employed. The µfluorescence analyses have revealed that Sr diffusion across the electrolyte interface occur mainly during electrolysis operation; whereas the process is very limited in fuel cell mode. This result was found consistent with SEM observations of a secondary Sr-rich phase after electrolysis operation. TEM-EDX and the X-ray µdiffraction reconstructions have allowed identifying this secondary phase as SrZrO₃. Based on these characterizations, the cell polarization curves and the local quantities (concentration of vacancies, adsorbates, etc.) within the O₂ electrode have been computed in both operating modes. The simulations
have shown that the electrolysis operation leads to a strong depletion of oxygen vacancies in LSCF material. It has been proposed that the depletion in oxygen vacancies under electrolysis polarization could drive the Sr release from the structure, and in turn, could explain the experimental results. A new mechanism of LSCF destabilization and SrZrO3 formation has then been detailed.

6. Cell degradation in electrolysis versus fuel cell modes
6.1 Steady-state “lightweight” stack model
For system modelling and techno-economic analyses, a “lightweight” steady-state solid-oxide electrolyser stack model has been developed and implemented in the Aspen Plus software package. The model has been designed in such way that it can be adjustable for both the steam electrolysis and co-electrolysis, and for both calculating the process conditions of the 3 kW prototype (built in WP5) and the MW-scale facilities (evaluated in WP2). The model inputs are the current density, the cell surface area, the number of cells, the area specific resistance (ASR), the Faradaic and heat losses, the input flow rates, the temperature and pressure. The ASR is expressed through a phenomenological polynomial equation calibrated on experiments. The model simulation provides as results the flow rates, the composition and temperature of gases the stack outlet, the operating voltage and the input electrical power required for the SOEC operation. During the project, the model has been validated and calibrated using experimental data from CEA and VTT (WP4). The validated version of the Aspen stack model was then provided to WP2 and used in the “sun-to-power-to-X system modelling” framework for the techno-economic analyses conducted by ENGIE and EPFL (WP2).

6.2 Dynamic stack model
As the SOEC is usually coupled with an intermittent electricity source, it was important to develop a dynamic model able to analyze the stack transient behavior. For this purpose, a time-dependent stack model has been built and implemented in Comsol multiphysics. The geometry of the stack used in the model corresponds to a simplified geometry of the 25-cell CEA stack prototype that was used in the SOEC system developed in WP6. Simulations have been carried out to investigate the stack thermal behavior under miscellaneous cases of current or air flow loadings. It was found that the model reacts qualitatively correctly, with a temperature increases for an exothermal operation point with the hottest spot located between the flow outlets (for a cross-flow stack configuration). As expected, it has been found that the average cell temperature drops to 710 °C in 2000 s when the current is set to a very endothermic operation. The analyses of these simulating results have highlighted the challenge to control the average cell temperature of a stack in endothermic operation with the anode air flow rate and its inlet temperature. In addition, the simulations suggest that transition from 0 A/cm² to a current corresponding to the thermoneutral voltage could cause more thermal stress in the cell in the case of the ramp rate is so slow that the cell temperature would have time to change significantly.

The main achievements is given below as a list of ‘highlights’:
→ Several three-dimensional reconstructions have been obtained on the pristine cell and after operation.
→ A new concept of accessible TPBs in the 3D microstructure has been proposed and used on the Ni-YSZ electrode operated in fuel cell and electrolysis modes.
→ A substantial Ni coarsening has been measured on the 3D reconstructions after cell operation. A power-law model was fitted on the data and implemented in the multiscale mode: the Ni coarsening explains between 20 to 30% of the cell performances degradation.
→ It has been found that the LSCF destabilization and the reactivity with the electrolyte is strongly
promoted under electrolysis mode. Thanks to the model, a mechanism of LSCF demixing driven by the electrode polarization has been proposed.

→ A LSCF and LSCF-CGO micro-scale model has been developed and implemented in the numerical tool for electrolysis and fuel cell mode. Once the reactive pathway validated, the role of electrode microstructure on cell performances has been investigated for cell optimization purpose.

→ A SRU model has been adapted to take into account the effect of pressure in steam and co-electrolysis. Once validated, the impact of pressure on the cell operation has been studied. A set of simulations has also been carried out to identify the best operating conditions under pressure.

→ A “lightweight” steady-state solid-oxide electrolyser stack model has been developed and transferred to WP2.

→ A time-dependent stack model has been built and used to investigate the stack thermal behavior under various cases of current or air flow loadings.

WP4: High Temperature Electrolyser Stack development

WP4 deals with the development and testing of SOE cells and stacks in both steam- and co-electrolysis mode operating at atmospheric and pressurized conditions. It comprises 5 tasks having their own specific objectives.

WT4.1 Test and characterisation protocols

Relevant test protocols have been established for cell and stack testing, both in electrolysis and co-electrolysis under varying pressure conditions. Besides performance mapping, the protocol also addresses durability tests under steady-state and dynamic variation conditions.

WT4.2 Development of optimized cells for SOE and co-SOE

This task includes pressurized operation and up-scaling to large area cells.

Following the suggestions of WP3 based on the conclusions of the modelling activities, the State-of-the-Art cells were optimized for SOE operation by improving the dispersion of GDC and LSCF phases in the oxygen electrode functional layer. This improvement was achieved by adjusting the particle size distribution of the powders used for the electrode fabrication and optimizing the ink production process. In particular, the dispersion of the powders, previously obtained by a conventional ball milling process, was processed by a more efficient Triple Roll Milling equipment. The resulting microstructural changes allowed to improve the cell performance 10-15%.

In addition, the chemical composition of the electrodes was modified by the addition of dopants to improve the adhesion on the diffusion barrier layer (DBL). Furthermore, the ohmic resistance of the cells was strongly reduced by reducing the thickness of the DBL. This result was achieved by modifying the viscosity of the DBL ink and optimizing the parameters of the screen printing process.

Finally, a further improvement on the performance and stability of the DBL was obtained within the frame of the parallel project Endurance and consisted in the addition of reactive elements to the Gd2O3-CeO2 phase.

WT4.3 Development of stack for SOE and co-SOE applications.

This task follows two distinct objectives: 1) the development and prototyping of a 1-3kW stack for pressurized operation up to 15bar (CEA), to be integrated in the solar-SOEC system (WP6-7); 2) the development of a large area stack design (>200cm2) aiming at the 10kW-class.
Pressurized stack development

A 25 repeat unit stack (3kWe), previously developed and tested by CEA for steam electrolysis and co-electrolysis at atmospheric pressure, has been adapted to SOLIDpower cells and to pressurized electrolysis operation for the project. The main development focused on the self-clamping system for stack tightening, which enables the stack to be placed in a pressurized vessel, thereby allowing operation under pressure (up to 15 bars) without applying a pressure difference between the inside and the outside of the stack ($\Delta P \leq 50$ mbar) (cf. results in WP6 and WP7). In addition, the contact elements and sealing concept have been optimized for SOPHIA cells and validated in several 1-cell stacks. It was shown that at atmospheric pressure, the cell and stack can be operated at high current density ($\Delta i \geq 0.6$ A/cm$^2$) even at 700°C, which might help in ageing resistance. Furthermore, these performance results were very close to single cells tested in the same conditions. This very low scattering observed between single cells (3 cm$^2$ of active area) and 1-cell stack (100 cm$^2$) validated the design of the stack in terms of electrical contact solutions. It must be also noticed that the limiting current is detected only at high steam conversion rate (90%), showing that the cell and stack are both able to operate at high steam conversion rates and consequently validating the gas flow distribution in the CEA design in these gas conditions.

Two SOPHIA 25 cells-stacks have been built for the project. The first stack was not sufficiently gas-tight to be operated under pressure. This was due to a bad positioning of the cells and/or Ni grids resulting from the important warpage of the cells. This issue was solved by positioning the cells with glue, thereby allowing the successful assembly of a second 25-cells stack that has been integrated in the SOPHIA system for pressurized tests in SOEC mode. Its performances after conditioning step at CEA were:
- stack tightness higher than 95%,
- operation close to thermoneutral voltage (32 V) at 700°C and -60 A (-0.6 A/cm$^2$) using nitrogen instead of air as sweeping gas in the anode compartment to protect the sealing performed during few tens of hours at atmospheric pressure. The same operating conditions have been proposed for the SOPHIA prototype at high pressure.

Large area stack development

A new design was developed by HTceramix based on SOLIDpower’s stack technology but with a larger active area (320cm$^2$). This also required the parallel development of a dedicated test-bench. In order to fulfil milestone MS4 (Proof of concept of large area stack operating in co-electrolysis), a 3 layer short-stack was assembled, corresponding to 960cm$^2$ in total active area. It was first qualified in SOFC mode for direct comparison with the SoA stack design SOLIDpower. The large area short stack successfully reproduced the nominal performance of a SoA SOFC stack design (>0.8V/RU at 128A, 0.4 A/cm$^2$, 750°C, 80% FU), indicating that no loss of performance accompanied the up-scaling. This is an important achievement, which brings us closer to 10kW-class stack manufacturing. The short-stack was then tested in steam- and co-electrolysis mode under different H2O-CO2 compositions, thereby fulfilling the proof of concept milestone (MS4).

In steam-electrolysis mode, thermo-neutral voltage (~1.3V) is reached at -208A (-0.65A/cm$^2$ 750°C). Increasing the CO2 content in co-electrolysis mode tends to increase the voltage at the same current, thereby lowering the performance, in particular at high current where gas transport becomes limiting. The large area short stack was then tested in steady-state conditions in steam-electrolysis mode at -0.75A/cm$^2$ (49% conversion) for more than 1100h without apparent degradation.

WT4.4 Cell and stack characterization at atmospheric pressure for SOE and co-SOE applications.
The goal is first to evaluate the performance of SoA cells and stacks according to the operating conditions defined in T4.1. This is used to set a benchmark for monitoring the progress. Stack will also be tested under dynamic load variation to mimic the coupling of SOEC with intermittent power sources (solar, wind).

Single cell testing
A detailed study of degradation mechanisms based on single cells testing was performed by CEA. It highlights that in similar operating conditions, the degradation rates were found to be higher in electrolysis than in fuel cell operation. To explain such results, both Ni agglomeration and LSCF destabilization have been investigated by coupling advanced post-test characterizations and modelling. Thanks to 3D electrode reconstructions, it has been found that Ni coarsening upon operation is not sensitive to the polarization, even if the agglomeration explains a significant part of the cell degradation. On the contrary, post-test analyses have revealed that Sr diffusion and formation of SrZrO3 at YSZ/CGO interface occur mainly during electrolysis operation, whereas the process is very limited in fuel cell mode. Therefore, the higher degradation rates measured in electrolysis mode could be related to the higher rate of LSCF demixing.

Short-stack testing
During the SOPHIA project, 7 short-stacks from SOLIDpower have been tested by VTT and EPFL. A test matrix that has been used to explore different operating conditions (temperature, gas compositions) and type of test conducted within the project (durability, load cycling). Altogether, more than 13000 hours of testing has been reached. The table also shows which gas compositions were investigated (SE=steam electrolysis, C1-C3=co-electrolysis with resp. 10, 25 and 45% CO2) and the type of stack. SoA refers to the state-of-the–art cell and stack technology from SOLIDpower. The improvement conducted on cells and stack components within T4.2 have been detailed above. Below, we will summarize the main results and try to extract the trends from this large amount of data.

Steady-state degradation
The durability test on stacks 1, 5 and 6 were performed at steady conditions and showed no overall apparent degradation during the test duration but instead a small improvement. However, EIS measurements performed periodically showed an increase of the ohmic resistance with time that is masked by a simultaneous improvement of the polarisation resistance. With stacks 2 and 3, the operating conditions were varied in consecutive sequences of 300h in order to check the sensitivity of the degradation rate towards different parameters such as current density, gas composition and temperature. The degradation rates varied between -17mV/kh (improvement) to +102mV/kh depending on the operating conditions. Despite some scattering, the following trends could be drawn for the apparent degradation rate:
• it decreases with increasing temperature;
• it increases with increasing current density;
• it increases with CO2 fraction once it exceeds 25%.
It should however be emphasized that the apparent degradation rate is strongly influenced by the temperature dependence of the overpotentials. As the overpotentials are usually smaller at high temperature than at low temperature, the voltage degradation rates will appear smaller at higher temperature than at lower temperature. As a consequence it is necessary to perform periodical iV measurements at a common reference temperature to quantify the real degradation rate of the cell/stack.
Post-test examination was carried-on at EPFL on a SOLIDpower short-stack that had been tested for more than 10700 h in steam electrolysis at 700°C within the frame of a former EU-project (ADEL). The following changes in microstructure were observed:

- Ni depletion at the interface with the electrolyte (to a depth of 5 µm).
- void formation in the electrolyte along and inside the YSZ grain boundaries, especially in proximity of the GDC layer
- formation of a dense mixed YSZ-GDC layer at the interface between electrolyte and compatibility layer, with the consequent formation of SrZrO3

Ni depletion and the accumulation of voids in the electrolyte created an overall weakening of the structure to cause the possible initiation of cracks. These alterations are expected to lead to a decrease in performance, lowering the available TPB sites in the cathode and obstructing the passage of oxygen ions because of the accumulation of SrZrO3 at the YSZ/GDC interface. These observations could explain the increase in the ohmic resistance observed by EIS measurements during the durability tests.

Degradation during load cycling

Stack 4 and 7 were tested in dynamic mode in order to identify additional degradation resulting from the rapid change of the current in relation with the use of intermittent power sources. Different load cycles were considered between OCV and the operating voltage (thermoneutral or exothermic) by varying the current ramping (3-1000 A/min) and the duration of the plateaus (30-600 s). Both steam- and co-electrolysis were investigated. The following conclusions can be drawn:

- Load cycling increases the degradation rate compared to steady-state operation but to a limited extent, suggesting that coupling the SOEC with intermittent power sources is feasible;
- The way load cycling is performed (i.e. frequency, plateaus duration, operating voltage and ramping rates) influences the degradation rate;
- EIS measurements performed before and after each cycling sequence showed that the additional degradation corresponds to an increase of the resistance attributed to cathodic charge transfer.

WT4.5 Cell and stack characterization under pressure (up to 15bar) for SOE and co-SOE applications. The objectives of this task are testing under atmospheric and under pressure and in steady-state conditions.

SoA single cells characterizations have been performed at CEA under pressurized conditions (up to 10 bar) in SOEC and co-SOEC using a specifically designed test-rig. Pressurization brings significant performance improvement at higher current density (ΔiΔ>1A/cm2) compared to atmospheric operation: syngas production is increased by 25% in the tested conditions of figure 3. This mainly results from a decrease of the concentration overpotential that becomes dominant at high current. These pressurized tests have been performed both in steam- and co-electrolysis modes. To our knowledge, it is the first experimental study reporting pressurized co-electrolysis. Durability tests were also performed under pressurized conditions for both modes. In co-electrolysis mode (at 3.7bar 750°C, 35% conversion), after a strong initial increase of the voltage during the first 150h, the degradation stabilized around +20mV/kh over 1500h. In steam electrolysis mode (at 5bar, 800°C, 50% conversion), the degradation rate was between 40 and 100 mV/kh, which is close to that measured at atmospheric pressure. EIS measurement showed that the degradation was mainly ohmic, indicating that the pressure does not significantly modify the degradation mechanism.
Main achievements (highlights)

• Successful implementation of microstructure engineering suggested by modelling results.
• 25 cells stacks with self-clamping system for operation up to 15bar build and integrated in system prototype.
• Proof of concept for large area stack (320cm²/layer) in co-electrolysis successfully demonstrated, followed by >1000h of steady-state operation without degradation.
• Detailed study of degradation mechanisms based on single cells testing.
• Intensive short-stack testing (7 stacks, >13000h). 3 steady state durability tests showed no apparent degradation.
• Durability under load-cycling conditions demonstrated with acceptable degradation.
• Cells have been tested under pressurized (up to 10bar) conditions in steam- and co-electrolysis mode.
• Co-electrolysis results under pressurized conditions reported for the first time.

WP5: System design and integration of energy source

Main objectives of WP5 are:

• Set up and refinement of flow chart for the prototype operation as well as for a scale-up plant
• Identification of optimal integration schemes of the pressurized high temperature co-electrolyser with a solar energy source

WT5.1 System Process Flow chart – Modelling and Simulation

Flow charts of the coupling of the CSP technologies to the steam electrolysis have been developed by DLR for a scaled-up hydrogen production plant, especially for a Direct Steam Generation plant, a pressurized air solar plant and for a Molten Salt Power plant. The simulations of these three solar plants have been carried out for a hydrogen production rate of 400 kg/h corresponding to a mobility scenario and for a hydrogen production rate of 4000 kg/h, which corresponds to an industrial process.

The process efficiency has been calculated for each case.

The performance of solar energy plants strongly depends on the site where they are erected and the corresponding insolation conditions. The annual average solar efficiency was calculated for each case studied.

Schematics for the experimental layout of the prototype experiment have been also produced. Data acquisition for the main parameters of pressure, temperature profiles, parallel and counterflow monitoring as well as the control loops and safety mechanisms for high temperature have been implemented in a prototype Process & Instrumentation Diagram, as well as a process flowsheet for the prototype, with further input by HyGear and a safety analysis by VTT. Three process flow diagrams (PFDs) have been proposed and analysed: Base, Sweep, and Sweep & recycling.

In the base flow sheet pure oxygen is produced at the anode, whereas in the sweep one, use of sweeping air at the anode side has been considered, for safety reasons and to avoid corrosion issues, related to the production of pure oxygen. The latter configuration allows mitigating the O₂ content in anode exhaust and allows also a more stable control of the system.

The sweep & recycling flow sheet has been analysed because the overall conversion as well as the stack efficiency can be increased by recycling the outlet product stream from the cathode side. However, the
compression of the recycled stream is required, introducing an energy penalty. As an alternative to a compressor, the application of an ejector has been proposed. However, taking into account factors as safety, controllability, manufacturing and handling, the sweep configuration has been proposed to be developed for the prototype. Superheated stream, slightly above the saturation temperature, avoids the partial steam condensation when the cold hydrogen from cylinder is added. A small hydrogen content (H₂ 10% vol.) is in fact necessary to limit the degradation of cathode material during transient operation. A compressor, placed upstream the SOE stack, provides the compressed air (up to 15 bara) required for the sweep stream fed to the cathode and the purging stream fed to the vessel. The sweep air flow rate has been defined to adjust the outlet oxygen content in the anode side to 50%. The purging air flow rate has been calculated on the basis of possible stake leakages.

The possibility to recover the heat from anode and cathode outlet streams has been considered. For this purpose, steam and air heat recuperators have been implemented in the process flow diagram. An outlet temperature around 600°C is expected for both anode and cathode streams. Due to the thermo-neutral operation, the final heating step, up to the operating temperature, is done by electrical heaters allowing also a better control and tuning of the outlet temperature. A steam conversion of 50% has been assumed. The cooled O₂ enriched air and hydrogen outlet streams are vented.

Material and energy balances for the proposed flow sheets have been performed. The heat duties for each heater and heat exchanger have been also evaluated.

A SOEC stack model was developed in Aspen modelling environment by VTT in strong relation with WP3. As a first step a steady state SOEC stack model was developed. The steady state stack model was to be exploited in WP5 in the system process flow chart of the prototype. This model was aimed to be combined into the process flowchart of the system in order to be able to analyse the operation of the prototype. This model was then updated based on experimental data obtained by VTT and CEA. In this model the effect of pressure can be defined. Faradic loss and heat loss can be defined for the stack. This version included some estimated SOEC input parameters of the prototype attained from the CEA. In addition, a recycling stream from produced syngas to the stack input was included in the model to ensure the correct iteration procedure of the energy balance calculations. The model was validated and calibrated using the data from WP4. The updated stack model was shared with partners and was used in sun-to-power-to-X system modelling in WP2 for the techno-economic analysis.

WT5.2 Component specifications, mechanical design & controls
On the basis of the process flow diagram and P&ID the specifications of the needed process equipment and instrumentations for the HTE prototype have been defined.

The stack is contained in a pressurized vessel. In order to maintain the same pressure and temperature in the stack and pressurized vessel, an air inlet into the pressure vessel is included. Moreover, a purging system of the pressure vessel is required in case of leakage from the stack; thus, an air circulation/purging circuit has been designed. The air stream lines are afterward connected to get the same pressure between the vessel and anode side. The pressure control between cathode and anode compartment has been also defined. Basically, the pressure regulation is performed by applying two back pressure regulators with a control loop. The heat recovery from both anode and cathode streams has been analysed. For the air heat recover, a three fluid counter-current heat exchanger is integrated. In particular, the hot air coming out from the anode and vessel is used to heat up the two cold air inlet, the sweep and purging streams, respectively. Similarly, the heat from the cathode side is also recovered by heat exchanging; the final stack inlet streams super heating, up to 750°C, it is performed by applying electrical
heaters. The P&ID also includes the needed units for the prototype testing when the system is not connected to the steam solar generator (i.e. at HyGear site). High pressure (up to 15 bar) steam generator and successive steam super heater have been integrated.

Moreover, the different operating pressure expected during testing at DLR site (max. pressure up to 3-4 bara) likely will require some instruments in the P&ID to double (one for testing at high pressure and other one for low pressure). The self-clamping part of the stack, i.e. an autonomous equipment for tightening, has been validated by CEA in terms of gas tightness and contact resistance.

WT5.3 Integration of the solar energy source

The following energy sources were considered by DLR for the coupling to a large scale electrolysis process: Molten salt solar tower, Direct steam generation in central receiver system, Pressurized air solar receiver, Parabolic Dish, Parabolic Trough, and Linear Fresnel. The solar tower technology working with molten salt as fluid leads to a higher annual capacity due the long term thermal storage system, which has been integrated and already demonstrated in these plants. It also enables to feed the electrolyser with a more stable steam flow, which is also an advantage for this technology. As the stability of the operation conditions – in particularly concerning the operation temperature - and the possibility of long term thermal storage are important factors for the operation of the pressurized high temperature electrolyser, it was finally decided to focus on the molten salt solar tower technology and the results of this task were also used in WP2.

In order to investigate the best solar energy partition (solar electricity versus solar heat) for a fully solar-driven high-temperature electrolysis (HTE) system, a variety of solar HTE system design were designed and studied. Particularly, EPFL proposed the used photovoltaic (PV) technologies for the electricity generation. EPFL developed a thermo-economic model of solar-driven HT electrolysis systems using pure thermal, pure electrical, and hybrid approaches. The model allows for a direct comparison of the three approaches using performance criteria (e.g. solar-to-fuel efficiency) and economic criteria (e.g. hydrogen cost) under various design and operational conditions, and under various material and device choices. For solar-driven approaches, three possible system approaches can be sketched using two relatively mature technologies:

i) thermal approaches using concentrated solar technologies to provide heat and to generate electricity through a traditional heat-driven thermodynamic cycle,

ii) electrical approaches using photovoltaic technologies to provide electricity and to generate heat through electrical heaters,

iii) hybrid approaches utilizing concentrated solar technologies and photovoltaics to provide heat and electricity, respectively.

Based on the proposed techno-economic model, a detailed analysis to evaluate both energy and economic performance for the proposed three systems was conducted under various design and operational conditions. Various operation conditions were simulated. Generally speaking, system (a) was able to access higher efficiency values while exhibiting higher production cost due to the more expensive costs for concentrated solar power systems compared to PV systems. Heating by electrical heater driven by electricity from PV (system (b)) showed only a solar-to-heat efficiency of about 13.5%. System (c) is the hybrid system which has both advantages: high heating efficiency of system (a), and cost advantage of system (b). This feature leads to a comparatively high system efficiency and potentially low hydrogen cost. For system (a) and (c), the low efficiency limit is about 6% which is absolute 2.5% higher than for system (b). This resulted from the higher solar-to-heat efficiency of system (a) and (c) using concentrated solar
technologies.

WP6: Prototyping & Factory Acceptance Test
Main objectives of WP6 are:
• Build high pressure (co-SOE) stacks
• Build and test all modules of the prototype
• Assembly (co-)SOE prototype system
• Factory Acceptance Test of prototype

WT6.1 Build and FAT stacks
Stack description and operation recommendations
A stack has been previously developed and tested by CEA for steam electrolysis and co-electrolysis at atmospheric pressure. [Di Iorio et al., SOE stack activities at CEA, EFCF proceedings, 2014]. This stack design has been adapted to (co-)electrolysis operation under pressure for the SOPHIA project: thanks to a self-tightening system and a strategy for pressure regulation, the stack can be placed in a pressurized vessel, allowing an operation under pressure up to 15 bars. In more detail, to facilitate the integration of the stack in the pressurized vessel and to optimize the sealing and the electrical contact, a mechanical loading is applied on the stack via an integrated self-clamping system. Furthermore, during transient and steady-state operations, the pressure differentials between the anodic and cathodic compartments and between the stack and the vessel are regulated at 50 mbar max.

Considering the stack instrumentation, the stack was delivered with voltage probes spot-welded to each interconnect to measure the voltage of each cell and two current rods fixed to the end plates. Holes have been drilled in the thick end plates of the stack to place thermocouples. The stack was delivered with 4 tubes, i.e. 2 inlet/outlet tubes for anodic and cathodic compartments, welded on the bottom plate. Note also that the bottom plate (O2 side) is ground connected.

The operating mode recommended to operate the stack under pressure was a “global” thermo-neutral mode at 700°C, i.e. a mean voltage per cell of 1.3V or else a current density close to -0.6 A/cm², with a steam conversion of 50%.

Manufacturing of stack(s) at CEA
During SOPHIA project, two SOPHIA 25 cell-stacks have been built for the project (“stacks 1 and 3”, one of which (“stack 3”) has been integrated to the SOPHIA system for pressurized tests in SOEC mode). In addition to these two stacks, a “dummy” stack, i.e. a second-hand and degraded stack, previously tested at CEA has been delivered to Hygear to validate the complete system before integrating the real stack number 3.

Stack 1:
A first stack has been manufactured by integrating SOLIDpower cells in 2016. However, after the heating up for sealing crystallization, leaks in cathodic and anodic compartments have been detected (through the outlet nitrogen flow rates measurements). The stack was then cooled and a repair of the sealing attempted. After a second heating up of the stack, the stack tightness was close to 95% in N2. The reduction of the H2 electrode has been performed by replacing gradually nitrogen by pure H2 (with N2 flowing in the other compartment due to the remaining H2 leakage). The quality of the stack was checked again but only 80% of H2 was recovered at the H2 outlet, mainly due to Repeat Unit n°18 and 19 (close to
the top of the stack). In spite of this problem, the stack was switched to electrolysis mode and performance i-V curves have been measured.

Initial performances of the stack were good (mean stack voltage close to 1.3 V at -1 A/cm²), however leaks detected in H2 and O2 compartments from inside to outside of the stack prevented the operation of this stack in pressurized conditions.

Stack 2 (also called “dummy” stack)
Waiting to build another stack integrating new SOLIDpower cells, a “dummy” stack, i.e. a second-hand and degraded stack, previously tested at CEA has been delivered to HyGear to validate the complete system.
This stack was delivered with three gas tubes: 2 inlet/outlet tubes for H2 compartment and only 1 inlet tube for O2 compartment, the O2 outlet tube having been broken during the stack dismounting at CEA following the previous test. That means that O2 outlet was evacuated directly in the vessel and that the system had to be adapted to accommodate this stack. This stack was tight: with 45g/h of H2 and 386 g/h of air, OCVs were between 1.115 and 1.125 V and P(H2) and P(O2) were about 20 and 50 mbar respectively. These values are consistent with data collected on previous tight stacks in the same gas compositions and flowrates.

Stack 3
The last stack (stack 3) manufactured for SOPHIA project has been manufactured in April 2017 and then delivered to HyGear for its implementation in the SOPHIA prototype. Cells integrated in this stack have been provided by SOLIDpower (SOPHIA 1G cell).
The stack 3 was characterized in terms of performances and tightness.

Instrumentation:
26 potential leads were initially spot-welded to the interconnect plates. However, 2 wires have been broken during the dismounting of the stack from the test rig (wires 11 and 14). The stack 3 was delivered with 4 tubes (i.e. 2 inlet/outlet tubes for H2 compartment and 2 inlet/outlet tubes for O2 compartment). The bottom plate (O2 side) is ground connected via the O2 tubes.

A specific heating-up procedure has been developed including gas tightness checks. Followed by an extensive performance verification with a j-V characterization and stationary operation. After cooling down the stack using a dedicated procedure the tightness of the stack was checked again and a complete leak-tightness report was made.

WT5.2 Build & test modules (steam generator, heat exchanger)
At HyGear all system components, except the stack and solar interface with steam generator have been build. Separately the steam generator needed for stand-alone testing at HyGear’s premises has been built as well as a small system which has been used to test the interaction of solar steam generator are the pressure control.

The steam generator was build according to the specifications defined in WP5. It is an electrically heated film-type evaporator capable of producing about 2 kg/hr of superheated steam at 220 oC and 15 bar.
Prior to shipping the system a sub-system prototype has been shipped to DLR. The goal of this prototype was to verify the differential pressure operation, check the interfaces between the HyGear system and DLR system.

The experiments with the sub-system fed with steam from the solar-steam generator showed initially an unstable behaviour of the differential pressure control, caused by a continuous cycle of condensation and evaporation of steam and water. Therefore, it was decided to alter the control logic by controlling air flow instead of the steam flow. The pressure difference as measured during start-up showed a clear stable value. The sub-system experiments have proven that steam from the solar simulator can be pressurized with the HyGear system and guided through the system, while maintaining proper (superheated) steam temperatures. More importantly, the control of the pressure difference was validated.

WT5.3 Build & FAT prototype

The system components, built in WP5, are assembled into the full system. Two different stacks were used in the system. The first stack had a broken anode outlet pipe for which adjustments had to be made in the interior of the vessel. The system with this stack installed was used for experiments at DLR. The second stack was used for testing at high pressure at HyGear. Each time prior to installing the stack the system was tested for leak tightness and the system was heated up. After the experiments at DLR additional heaters were installed with a higher power and additional insulation was put around the components to minimize the heat losses.

WT5.4 Safety and risk assessment

The work on the safety and Risk assessment is divided in three parts. The first part consists of a safety study of the pressurized high-temperature electrolyser prototype being developed in SOPHIA project. The second part consist of a review of the applicable EU directives that are applicable to such pressurized high-temperature electrolyser. The third part consists of a quantitative evaluation of the consequences of the most significant identified hazard. The event corresponds to the full rupture of the hydrogen line inside the container in which the system is located.

Pressurized high temperature electrolyser system – Review of applicable EU directives

The aim of the review was to identify and justify which safety related EU directives must be complied in the design and/or manufacturing of pressurized high-temperature electrolyser systems (based on SOEC technology for example).

Pressure Equipment Directive (PED), Low Voltage Directive (LVD), and Electromagnetic Compatibility (EMC) Directive are clearly directives that must be complied in the design and/or manufacturing of pressurized high-temperature electrolyser systems. Scope of Simple Pressure Vessel Directive (SPVD) is very limited and it is probable that it will not be applied. Machinery Directive (MD) is not applicable to pressurized, high-temperature electrolyser itself, because according to MD an electrolyser is not a machine. However in the entire system there are single equipment (e.g. pumps, compressors...) for which the requirements of the MD must be followed.

ATEX Equipment directive shall not be applied to the whole system, but there may be certain areas in the system where it must be applied. ATEX Workplace Directive sets requirements for operation of the system - not actually for design or manufacturing. Also requirement of Seveso directive and REACH are mostly related to operation phase of a system.
Quantitative analysis of the major risk scenario: explosion of a 10-ft container

Introduction

The quantitative analysis that was carried out in 4 followed the guidelines given by The Finnish Safety and Chemicals Agency (Tukes). The guidelines give info on how various accident scenarios need to be taken into account when citing production facilities.

The scenario investigated here is a complete break of a hydrogen feed line to the vessel. Hydrogen from the pipe keeps flowing to the container as long as the pressure in the hydrogen tanks is larger than the pressure of the pressure reducer. The resulting flammable mixture ignites leading to a deflagration. We looked into

1. Closed vessel deflagration: Assume that the container is completely sealed. Deflagration raises the internal pressure of the container to a very high level and the container bursts.
2. Vented deflagration: Assume that the container is open from one end (i.e. doors are open): What is the over-pressure inside the container and what is the over pressure of the external explosion.

The calculation methods used here are based on SFPE Handbook and CPR “Yellow book”, unless otherwise stated. When possible, the calculation results are compared with observations from real events.

Based on the calculations, it is evident that an explosive mixture may form in the container. Assuming that the doors work as explosion vents with activation pressure of seven kPa, the resulting flames will pose a threat up to 22 meters directly in front of the doors. The possible external explosion may pose limited structural damage up to 7.7 meters from the explosion. If the container doors are assumed to separate from the container at 8.4 kPa internal pressure, the doors may be propelled 65 meters from the container. Only deflagrations have been considered. Detonations are unlikely, since the energy needed to start a detonation is several orders of magnitude larger than that needed for deflagrations (106 J vs 10-4 J). It is also important to note that the probability of the events considered here has not been considered and numerous preventive measures are taken to avoid this scenario such as hydrogen detector in the container that commands automatic valve to isolate the hydrogen source from the container and sufficient ventilation to comply with ATEX zone 2NE inside the container.

WP7: System testing and analysis

Main objectives of WP7:
• To prove the principle of a solar-powered SOE system
• To prove the reliability and practicability of a SOE system integrated with a solar heat source
• To develop and establish the operational parameters as well as strategies and to determine the boundaries of operational range

WT7.1 Preparation of the solar interface

The Solar Interface coupling the equipment designed by the different partners, including the pressurized electrolyser, auxiliary heat exchangers and side components has been adapted to the experimental application of superheated steam production using solar-simulated power at the DLR’s Solar Simulator. The DLR’s high flux solar simulator is chosen due to easier -unbound to sun- operation. Furthermore, in for safety reasons, this room has the ability to put equipment with explosion risk on the outside (e.g. electrolyser). The radiative flux profile produced by the 10 lamps with 20 kW power has been adapted to a flat aperture profile at 60 cm from the window plane in front of the simulator, and several experiments have...
been carried out achieving above the required superheated steam flowrate for the stack integration. The focal point has been adjusted by thermal imaging and calibrated laser pointers, and a novel 3D-axis positioning system has been integrated into the Simulator Platform, along with a remote automatized interface. Auxiliary systems such as the monitoring control cabinet, steam condenser, frequency converters and data acquisition systems have been installed for the receiver experiments relevant to this project and have been continuously updated. Steam trap has been successfully added to the solar interface and tested in order to separate the overheated steam and condensate. An electro-pneumatic valve has been added to better control the pressure surges. An analogue balance has been also added in order to measure precisely the remaining condensate at the output of the receiver.

WT7.2 Modelling and optimization of the solar receiver
A detailed and validated numerical model for a cavity receiver with tubular absorbers was developed. The model coupled a 3-dimensional heat transfer cavity model (solving for conduction, natural convection, and radiation inside the cavity receiver) and a 1-dimensional two-phase flow model in the absorber tubes (solving for two-phase flow phenomena, specifically flow boiling inside the tubes). The model was validated with literature data first and then more specifically with the DLR experiments of the helical absorber tube (campaign on February 2016). Predicted temperatures along the absorber tube agreed very well (within 0.65%) with the experimentally measured temperatures (measured by thermocouples). The developed numerical framework provides a flexible and accurate tool for the performance evaluation and engineering design of indirectly irradiated solar cavity receivers for the direct stream generation. The numerical model was then exploited for guiding more practical and more efficient receiver designs for a next generation receiver development. Particularly, we focused on two different receiver designs (receiver 1: cavity receiver with helical absorber tube, and receiver 2: cavity receiver with straight tube absorbers). We investigated them as a function of various operation conditions, geometrical and dimensional variations, and for various material choices and properties.

Under reference conditions (water flow rate of 0.3 g/s, solar energy input of 1.5 kW, inlet pressure of 5 bar, and inlet water temperature of 303 K), receiver 1 exhibited higher re-radiation heat losses but smaller conductive and convective heat losses than receiver 2. The solar-to-thermal efficiency – defined as the fraction between the enthalpy change of heat transfer fluid (HTF) and the incident solar energy – was always higher for receiver 1 than receiver 2, due to higher and dominating conductive and convective heat losses for the latter. This inferiority in solar-to-thermal efficiency of receiver 2 was not observed when a larger solar power input was considered (for the same aperture size). At these elevated power levels, re-radiation heat losses dominated, which were generally higher for receiver 1. The flow rate had a significant effect on the solar-to-thermal efficiency and the fluid outlet conditions. In general, the efficiency increased with increasing flow rate for receiver 1. Receiver 2 showed an increasing efficiency with flow rate only when the flow rate was smaller than 1 g/s.

Receiver 1 was more sensitive to variations in pressure compared to receiver 2. In general, the solar-to-thermal efficiency decreased with increasing inlet fluid pressure. Under reference conditions, there existed a pressure range (10 bar-15 bar) where the decrease in efficiency was more significant. Changes in the component’s material showed some surprising changes in the observed trends. For example, a higher absorber tube surface emissivity always favored higher solar-to-thermal efficiency. However, receiver 2 showed larger solar-to-thermal efficiency than receiver 1 when the tube emissivity was smaller than 0.58 while receiver 1 was more efficient for larger emissivities. For receiver 1, the fluid inlet position played an important role in determining the solar-to-thermal efficiency
of the receiver. When the re-radiation heat loss dominated (at large input solar power, 10 kW in this study), the top inlet positioning (i.e. cold inlet fluid close to the aperture) showed better performance, resulting from reduced re-radiation losses when cold fluid passed through absorber tube parts close to the aperture. The bottom inlet showed to be the favorable option when the conductive and convective heat losses dominated (at small input solar power, 1.5 kW in this study).

In terms of dimensions, smaller tube diameters always led to higher receiver efficiencies. Receiver 1 exhibited higher efficiency than receiver 2 under reference conditions and when the inner tube diameter was smaller than ~7.5 mm. On the other hand, receiver 2 showed larger efficiency at inner tube diameter larger than ~7.5 mm.

The reference shape of the helical tube (cylindrical winding) of receiver 1 resulted in higher solar-to-thermal efficiencies than the two conical shapes (shape 1: a truncated cone with large circle close to the aperture, and shape 2: a truncated cone with small circle close to the aperture). Shape 1 showed very close performance to the reference case and the difference between the two can be reduced with a larger solar power input. A significant reduction in efficiency was observed in the case of shape 2 due to very high re-radiation losses, even more prominent at higher solar power input.

The developed tool is useful for the prediction and optimization of solar cavity receivers with tubular absorbers. Specifically, comparison of alternative designs, dimensions and arrangements, operating conditions, and material choices and properties were done and quantification of the performance and sensitivity to the performance were obtained.

In addition to the numerical model and optimization efforts at EPFL, a lab-scale 1.5 kW double helical tube receiver for the co-generation of high temperature steam and sweeping gas was designed and fabricated. The cost for the design and fabrication were provided by alternative sources. The two parallel absorber tubes were made of Inconel 600 with one helical tube for direct steam generation and one for sweep gas heating. Measurements demonstrated a solar-to-thermal efficiency of 69% at a solar input energy of 915 W, inlet water flow rate of 0.9 g/min, inlet N2 flow rate of 0.5 L/min, and inlet fluid temperatures of 298 K.

The receiver was designed in a way that an electrolyzer stack could be directly connected to the receiver tubes’ outlets for a direct feed of the stack in order to reduce transport losses. The electrolyzer stack was thermally insulated but incorporated in the back of the receiver cavity in order to form an integrated reactor. Thermal tests have shown that a stable heating rate of ~200 K/h can be achieved by gradually increasing the input power from the solar simulator and carefully adjusting the inlet flow rates. These experiments provide evidence that a pathway for a more integrated approach of solar-driven electrolysis is feasible and potentially advantageous in terms of efficiency.

WT7.3 Testing of solar interface
Comprehensive testing was performed by the DLR at the Solar Simulator in Cologne. Several campaigns for the study of superheated steam were carried out for the purpose of direct water to steam at high temperatures and a large body of data was collected and analysed. Two main technologies were compared, namely, an updated multi-tubular parallel flow solar receiver, and a novel, state of the art spiral multiphase receiver-reactor, ultimately choosing for the spiral reactor to serve as the solar interface with the electrolyser stack developed by the SOPHIA Partners. At first, the multi-tubular receiver was updated and enhanced for the purpose of evaporating water that would supply steam to the prototype electrolyser stack. The material selection, relevant pre-testing of different pressures and maximum flowrates with saturated steam were investigated. These results were finally put to work in the June 2015 Campaign, where the two technologies were compared to select the most suitable for water boiling. Experiments were
carried out at a slight over pressure of 1.3 bar (a). The flow rate was 5 ±0.1 kg/h, with a 1 kg/h increase
every 20 min, the maximum pressure achieved by 800°C steam was 4.5 bar at the outlet of the spiral and
inlet of the condenser. In summary, the design and habilitation of the novel spiral reactor represents a
huge increase in efficiency, a leap in technological advance for direct steam generation from liquid water,
and is deemed to be the most suitable technology for integration with the hydrogen production stack.
Several test campaigns were performed then with the novel developed and optimized spiral reactor for the
purpose of production of direct water to steam at high temperatures. Temperature and pressure sensors
were added to help picture the systems behavior and to determine a value of its efficiency.
The test campaign performed in February 2016 investigated the effect of heating bands and additional
insulation on the system’s performance. Also, the influences of the lamp set-up as well as the usage of a
cooling shield were regarded. The aim was to provide a homogenous and steady pressure and
temperature level for a potential electrolyzer stack. According to this campaign the temperature and
pressure levels required for the steam production were achieved.
The experimental campaign performed in June 2016 investigated the use of steam massflow equipment
and an electro-pneumatic valve to control the mass flow of the produced vapor. The experimental data
indicate an overall steady steam behavior. The pressure level fluctuates by 0.05 bar only. Also, the
temperatures on the spiral do not present any major fluctuation but depict a steady behavior. The mass
flow was set and regulated to 2 kg/h. The results indicated a proper control of the steam mass flow.
Finally a last test campaign of the solar interface took place at DLR on November 2016. In this campaign,
a “pre-system” delivered by HyGear and consisting of a number of valves, vessels and some sensors was
coupled to the solar interface in order to have some preliminary results. Before testing, cone insulation,
tracing and frame stability were improved. Leakage test had been performed and showed that the system
was tight. The process control system has been implemented in LabVIEW. The steam mass flow can
selectively be controlled manually or by a PID algorithm whereas the water mass flow is set by a simple
two-level-controller. The user is able to vary the setpoints for both controls during operation. The coupled
systems have been tested. Tests have been carried out with saturated steam and with superheated
steam.

Tests with saturated steam produced solar steam with a controlled massflow of 1.7 kg/h using 3 lamps of
the solar simulator to start up and then 2 lamps for the steady state operation. The temperature variations
are only minor and the temperature reaches about 150°C for the saturated steam temperature at 4 bar(a).
Regarding the pressure values no major deviations are to be seen. The fluctuation stayed within the limits
of 45mbar.

As saturated steam tends to condensate quicker than superheated steam, it was chosen to run the
following tests with higher temperatures and less tracing. Tracing was used only for the tubes that led out
of the solar simulator. The temperatures are significantly higher (350°C) than in previous tests. The
temperatures generally appear steady and only indicate minor fluctuation which not cause harm to the
system. Regarding the system pressure, no irregularities are visible. The level remains evenly at 4 bar(a)
and indicates a fluctuation of ca. 45mbar. The tests with superheated steam are easier to control and also
provide a higher yield of steam. Less condensation occurs and keeps the overall system more efficient with
regards to its steam conversion.

WT7.4 Installation and start-up of the overall system
Following the tests with the pre-system, some design changes have been done to improve the overall
performance of the system. First, the system should provide all its condensation traps before entering the steam flow measurement to determine the exact amount of steam entering the Hygear system. At best, the measurement takes place right before the electrolyzer stack. Also, the steam control valve has been put parallel to the steam measurement. Besides, the tube diameter has been reduced from 10 to 6 mm in order to increase the flow speed (which was about 6.4 m/s with a tube diameter of 10 mm). Therefore, less energy is absorbed by tubes or parts of the system and consequently less condensation is to be expected. Also, the lines have been designed as linear as possible. Last, more and smarter insulation has been used to reduce heat losses to the environment. The stack system has been delivered by HyGear in a container with integrated ventilation system in its roof which has been placed with a crane near from the solar simulator building in order to be connected with the solar system. During normal operation the container was closed, and air was sucked in via openings on the side and exiting the container via the vent on top of the container. In addition hydrogen sensors were located inside the container for safety reasons.

The tests were operated with a “dummy” stack (i.e. a second-hand and degraded stack, previously tested at CEA, see details in periodic report M42, WP6). The piping inside the pressure vessel needed to be adapted to accommodate this stack as the cathode outlet pipe was missing. For safety reasons both the cathode and the anode were flushed with nitrogen. The stack has been installed in the pressurized vessel and a pressurized test at 3 bar and room temperature (RT) has been performed, highlighting that the system was initially tight under pressurized conditions and RT. Then, the insulation of the vessel had been done. Tracing and heating wires have been provided by DLR for the tubes between the solar simulator and the electrolyzer system as well as rain protection material. Container openings for piping in/out are available and flanges have been used for connection. DLR took care of gas supply. The process control system has been installed and tested. Specific check lists and start-up procedures have been elaborated. The initial operation of the overall testing device has been done carefully and step by step. Finally first pressurized tests have been done.

WT7.5 Testing and evaluation of the overall system

The testing and evaluation of the system was done in two phases. In phase one the complete system was tested at DLR receiving steam from the solar simulator, the maximum operating pressure was limited to 4 bara; in the second phase the system was tested at HyGear using steam from the internal steam generator and operated at 15 bara.

Phase one
The electrolyser system was slowly heated up from temperature. However, after 24 hours the thermocouples located at the stack indicated temperatures in the range of 160°C. Therefore, it was decided to send superheated steam from the solar reactor to heat the stack but it was not enough to reach in the stack the minimal operation temperature of 650°C as the system was not designed for this purpose.

The gas temperatures at cathode and anode side, inside and outside the vessel, during the heating-up of the system are shown in the figures below. In about 40 hours the cathode gas temperature reaches a plateau of about 450°C; reducing the gas flow has some influence on the temperature. The temperature of the gas after the stack increases to a temperature of 250°C. The anode gas temperatures behave similarly. Due to experimental problems the temperature of the stack could not be measured. Based on the gas temperatures it is assumed that the temperature of the stack might have reached a maximum of 400°C.

The differential pressure control is capable of keeping the difference below 50 mbar even when large
pressure fluctuations occur.
Finally after having tried to solve the heating difficulty of the electrical heater, the stack was again heated up during 24 hours and superheated steam was sent to the electrolyser in order to produce hydrogen. Although the thermocouples did not indicate the required temperature for the stack, the electrolyser was operated and some hydrogen has been produced, 35 ml/min.
The coupling of a solar reactor producing steam and a Solid Oxide Electrolyser system has been successfully demonstrated. By solar energy superheated steam was produced and sent to the electrolyser system and hydrogen was produced.

Phase two
In the second phase the electrolyser system was tested in a stand-alone configuration in which steam is produced using an electrically heated steam generator. Prior to testing the system was adapted to reduce heat losses and to be able to provide more heat to the system. After the adaptations the functionality was again tested. Subsequently the final stack was built into the system. Under atmospheric pressure the system was heated up slowly. About 2.5 days the system was operated at a pressure of 3 bar(a), then the pressure was increased to 7.5 bar(a). Finally the system pressure was increased to 15 bar(a). During constant pressure operation the pressure difference between anode and cathode was about -5 mbar; only during pressure increase and decrease the difference increased occasionally to + or -50 mbar. This again shows that system control is able to limit the pressure difference below 50 mbar even at high operating pressures.

During two days the stack in the system was polarized, so sending a current to the stack in order to produce hydrogen and heat to increase the stack temperature. The latter was not successful, the stack temperature remained at about 450 °C. Nevertheless a maximum current of 1.2 A was observed, corresponding to about 200 ml/min.

WP8: Dissimination and Exploitation
The objectives of this work-package aim at maximizing the SOPHIA project impact in terms of electrolysis and co-electrolysis components, stack and system performance and efficiency at European level. The dissemination and exploitation activities will cover the following:
- Define an exploitation roadmap & business plan for technology upscaling and commercialisation taking advantage of both external stakeholders’ inputs and project achievements with exploitation potential.
- Manage communication outside the consortium in agreement with the CA by means of appropriated channels such as public website, workshops, conference communications, publications, etc.
- Inform the scientific and technical community as well as relevant networks and platforms about the results of the SOPHIA project and about their applications in high impact sectors (industry, energy, transport). This will be done by promoting and participating to conferences, congresses, industrial exhibitions, etc.
- Organize 2 workshops: i) one on the influence of operation conditions on stack component durability, with major attention given to pressure and CO2/H2O ratio ; ii) one on system designs, scaling up issues and deployment strategies.

WT8.1 Exploitation and Business Plan
During the second SOPHIA project period, ENGIE, with the support of consortium, has proceed with the
market assessment analysis, particularly on the potential market for renewable add-value products as hydrogen, methane, methanol and gasoline and in order to understand how these products can became competitive compared with the fossil ones.

Business models for each scenario and for two capacities production: 4,000 and 20,000 kgH2/d have been studied. In this deliverable “raw” production costs obtained in deliverable D2.5 will be integrated in an overall business case that consists on considering that the valorisation of all co-products are locally valorized. The considered “positive externalities” are:

- the valorisation of oxygen: a co-product of the electrolysis reaction,
- the valorisation of excess heat available and produced by CSP during the period where the sun irradiation is the highest and not integrated in the process. Two valorisation ways were considered for this “fatal” heat: (i) heat that is directly sold and injected on a local heat network or (ii) high temperature heat that can lead to supplementary production of added value products as hydrogen, methane, methanol, etc. which could lead to lower production costs due to CAPEX/OPEX mutualisation particularly for CSP.

Indeed, the additional CAPEX and OPEX required for this additional and occasional production are very low compared to CSP costs.

- the incorporation of a CO2 credits for emission avoided.

Market assessment

CPS potential development in Europe, expressed in installed capacity, will increase by 2040, depending on scenario from 8 to 14 GW.

Currently, early movers as Spain leads the European market of CSP with 2.3 GWe installed in the last five years. New investments are currently on hold but an upturn is expected. IEA forecasts an increasing of CSP installed capacity in the next few years leading in 2030 to an electricity capacity production between 4 and 6 GW according to the different scenarios.

Energy market analysis in Europe shows that today, the main fuels consumed in road transport are still diesel and gasoline. Other fuels, such as biodiesel, biogas and natural gas, represent only 5% of the total. However, they have an interesting potential in Europe.

Indeed, regulation will become more drastic: it enjoins, by 2020, a 10 % share for renewable energies in the transport sector. Moreover, fuel suppliers will have, by 2020, to reduce by at least 6% the life cycle GHG emissions per unit of energy of fuel used in the Union by road vehicle. This leads to the following potential outlook for 2020:

- A 2.5 % share for “advanced” biofuels (Biogas, Biomethane, Renewable Methane, Bio-methanol, Renewable Methanol, Renewable Electricity, Renewable Hydrogen, ...)
- A 7 % share for “conventional” biofuels (Bio-ethanol, bio-diesel).

“Advanced” biofuels could represent a huge market. If renewable hydrogen, methane, methanol or gasoline provided 100% of the « advanced biofuel », 2.5 % sub-target (6.9 Mtoe) in 2020 is respectively equivalent to: 2 Mt of hydrogen, 80 GWh renewable methane, 6.6 Mt of gasoline or to 12.7 Mt of methanol. Direct competitors of these “advanced fuels” are fossil fuels (gasoline, diesel, CNG) but also biofuels (bioethanol, bioCNG) and electricity for electric vehicles.

Moreover, renewable hydrogen, methane, methanol and gasoline will compete on “advanced” biofuel market. To evaluate the potential market, it will be considered, that each renewable fuels (hydrogen, methane, methanol and gasoline) could address 20% of this potential market. The 20% remaining could addressed to other renewable fuels as renewable electricity.
With this assumption, the annual market of the renewable fuels for mobility to the 2020 horizon are: (1) 0.4 Mt of renewable hydrogen, (2) 16 TWh of renewable methane, (3) 2.5 Mt of renewable methanol, (4) 1.3 Mt of renewable gasoline.

Considering these numbers and in order to identify the potential target on SOPHIA concept industrialization and commercialisation, Task 8.1 also aimed to quantify, in function of the market potential for these renewable fuels, the number of units that could be addressed to respond to the market needs. This exercise has been done supposing that SOPHIA concept market share is supposed to reach 20% for each different renewable fuel. The industrial potential deployment for renewable hydrogen market for mobility, depending on unit size ranges from 11 to 55.

Other market segments have also been identified for methane and methanol (in particular for industrial uses). A detailed state of the art is presented in deliverable 8.4.

According to main results obtained in WP 2 and WP8, through the different studies performed for each case study (defined in WP2) and its optimization through the integration of each case study in a particular business model aiming the overall valorization of all co-products and positive externalities of such Power to X chain, it is possible to conclude that:

Renewable hydrogen valorization for mobility use:
• CSP can produce, under adapted conditions of solar irradiation and for an important capacity production, competitive hydrogen for a mobility application particularly when the CSP plant is connected to power grid (even if limited to 30% of electricity supply).
• The competitiveness of hydrogen can be even more improved with the external valorisation of oxygen as a co-product of electrolysis and mainly heat.
• It is also possible to conclude that, cases considering CSP only source of electricity and heat can reach competitiveness for hydrogen production if SOPHIA concept is integrated in a well-established industrial network where co-products (and mail heat) can be almost totally externally valorised.
• Profitability can also be promoted by the increase of the production capacity. Increasing the production capacity of 4,000 kgH2/day to 20,000 kgH2/day can lead to a decrease of hydrogen production cost from 13.5 €/kg to 10.2 €/kg

Renewable methane, methanol and gasoline valorization:
SOPHIA concept does not seem to reach all required profitability conditions to assure a competitiveness for other final products as methane, methanol or gasoline, for the studied case studies, even if external valorisation of all co-products is taking into account. Quite important subsidies for CSP CAPEX are still required to achieve this competitiveness (from 55% to 85% according the cases).
This competitiveness could be eventually reached in other regions with higher DNI outside Europe. For these concepts, complementary R&D work on overall process optimization must proceed and in particular, on developing cost-effective technologies (for CSP heat and electricity production mainly but also for the thermo and electrochemical technologies). Synergies with other electricity sources need to be also studied.

WT8.2 Dissemination follow-up
Several articles on the part developments within the SOPHIA project have been published or submitted. Also several presentations have been given by the beneficiaries.
In addition a master thesis and a bachelor thesis were prepared on techno-economic analysis.

### WT8.3 Organization of dissemination/exploitation project workshop

A workshop "Degradation Mechanisms in Solid Oxide Cells and Systems" was held on February 17, 2017 in the Catalonia Hotel, in the center of the city of Barcelona. The workshop was co-organized by five FCH JU projects: ENDURANCE, DIAMOND, SOPHIA, SOCTESQA and ECo. The aim of the workshop was to ensure a platform for professional discussions and exchange of expertise in one of the most challenging issues in the commercialization of SOFC: degradation and durability. Different aspects covering cell components, seals and interconnectors, as well as stacks and systems were discussed and diverse approaches presented: from modeling and prediction of degradation, through experimental confirmation and observations to diagnostic tools for early detection and mitigation strategies.

The workshop gathered a mix of high-level researchers and industrial representatives involved in the development and market introduction of solid oxide cell systems. At the workshop an overview was given of the achievements of the SOPHIA project.

A final dissemination and exploitation workshop was held during the final meeting.

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**Potential Impact:**

### 4.1. Potential impact

The impact from the R&D done within the SOPHIA project can be divided in various categories:-

- Technical impact
- Commercial
- International
- Educational
- Integration in Europe
- Environmental

### Technical impacts

Within the project SOEC technology and a prototype system has been developed capable of producing hydrogen from water and solar energy. In more detail the following technical improvements were reached:

- A methodology for holotomography reconstructions has been developed;
- Microscopic models to macroscopic models have been developed;
- A new cell structure and an improved diffusion barrier yielding an improved performance;
- Cells for pressurised operation in both electrolysis and co-electrolysis have been developed;
- A self-clamping system for stacks has been developed;
- A solar receiver and steam generator has been developed able to produce 2 kg/hr. steam at temperatures over 200 oC and at a pressure of 3 bar ;
- A differential pressure control system for SOE systems has been developed;
- A complete 3 kWe high temperature electrolysis system capable of operation at a pressure of 15 bar has been developed.
Commercial impacts
The project was set up to demonstrate the capability of connecting a high temperature electrolysis system to a solar reactor producing steam, to investigate the market potential of such systems and to show the possibility of producing syngas by co-electrolysis of steam and CO2. On system level commercial impacts will arise only after a several years. Additional development and cost reductions are needed.

HyGear produces hydrogen generation systems. Currently hydrogen is produced using Steam Methane Reforming. The SOPHIA project has provided HyGear with the knowledge to widen its technology base for producing hydrogen in future years.

SP is a major developer and supplier of SOFC systems. It is constantly improving its cells, stacks, and systems. The results obtained in the SOPHIA project will assist SP in this effort.

International Impacts
The outcome of the project will help the partners within the project to compete on the international stage with SOEC systems and its components, and with steam generating solar receivers. The market for high temperature electrolysis systems is an international one. Large scale SOPHIA-like systems can be deployed in Southern Europe as the market analyses have shown. Deployment of stand-alone SOEC systems can be worldwide.

Educational Impacts
EPFL is an important institute for education, training and PhD students in the field of system modelling, solar receiver modelling and fuel cell and electrolyser research. The research institutes VTT, CEA, and DLR all involve master and PhD students in their research. During this project 1 bachelor student (DLR) and 1 master student (VTT) and 2 PhD students have worked on the project (EPFL, CEA)

Impact on Europe
The project has strengthened the European position in the field of Solar powered SOEC-systems, of cells and stacks development of academia, research institutes and SMEs/industry. Much knowledge has been gained on cell architecture, stack design and system design, but also in the field of markets and regulations. The RTD performers gained additional knowledge and experience in the field of SOEC and solar receivers. They will use this experience for future research and support of the industry. The SMEs have strengthened their position in Europe and the rest of the world.

Environmental and Economic Impacts
The technology developed can be used for producing hydrogen and syngas from renewable energy. Widespread adoption of SOEC technologies will give the environmental benefits of reduced fossil fuel usage (via increased efficiency) and reduced emissions.

4.2. Dissemination activities
The work in the project was mainly valorized and disseminated through 34 articles (cf. list of publications),
A workshop "Degradation Mechanisms in Solid Oxide Cells and Systems" was held on February 17, 2017 in the Catalonia Hotel, in the center of the city of Barcelona. The workshop was co-organized by five FCH JU projects: ENDURANCE, DIAMOND, SOPHIA, SOCTESQA and ECo. The aim of the workshop was to ensure a platform for professional discussions and exchange of expertise in one of the most challenging issues in the commercialization of SOFC: degradation and durability. Different aspects covering cell components, seals and interconnectors, as well as stacks and systems were discussed and diverse approaches presented: from modeling and prediction of degradation, through experimental confirmation and observations to diagnostic tools for early detection and mitigation strategies.

The workshop gathered a mix of high-level researchers and industrial representatives involved in the development and market introduction of solid oxide cell systems. At the workshop an overview was given of the achievements of the SOPHIA project.

A final dissemination and exploitation workshop was held during the final meeting.

4.3. Exploitation of results

The SOPHIA consortium can be divided into two partner groups, Industry and RTD-performers. Each group will identify the best type of exploitation action and the specific audience of the exploitation activities. Some of the exploitation activities will have direct impact on the technical improvements within a short time frame, while others will lead to future impact for the end users and technology manufacturers.

The industrial partners within SOPHIA will focus their exploitation activities on improving their current technology and business position in existing markets and on the creation of new markets beyond the markets addressed in SOPHIA. The industrial partners will use the technical improvements in a direct manner to shorten turn-around times to product readiness.

The exploitation goals of the RTD performers are different, yet complementary to those of the industrial partners. The technical improvements and developments will be integrated into the teaching courses but will also flow into the research activities of the RTD performers. For instance CEA’s business model consists in developing technologies for the industry and bringing them to market thanks to technology transfer to industry, either large industries, SME’s or start-ups.

This approach will lead to being more attractive for Ph.D. master and graduate level students to join the RTD performers. More strategically, graduates from academic partners will be trained for working in the related industry but also in related research work. The long-term pay-off of the RTD performers will lift SOPHIA’s technology being general part of teaching lessons.

As an example the methodology developed by EPFL in the frame of SOPHIA for the holotomography reconstructions (random displacement to correct the ring artifacts) can be applied not only for the SOFC electrodes but also for all kind of absorbent materials. It is already currently used on the beam line at ESRF for the characterization of different materials such as the components of batteries for example. All the models developed in the frame of SOPHIA will be also used for several forthcoming studies and projects dedicated to SOFC and SOEC. As an illustration, the O2 electrode model proposed in SOPHIA is currently used and extended in the FCH European INSIGHT project (Grant Agreement n° 735918) to detect the characteristic frequency evolution upon operation. It could provide a relevant tool for the
analysis of the fuel cell or electrolyser ‘state of health’. The pressurized co-electrolysis model is also used in the European ECO project (grant agreement n° 699892).

The exploitable results are listed below in groups, each group followed by short plans for exploitation.

Exploitable result: Cell and Stack
1 Improvement of the barrier layer by SP
2 Improvement of the oxygen electrode
3 Improved sealing design
4 Better steel protection
5 Strategy for pressurised stack operation
6 Large area stack developed
7 Microscopic to macroscopic models developed

Exploitation plans
1 The results developed within the SOPHIA project will be applied in the current SOFC technology by SP.
2
3
4
5 CEA has improved its leadership on SOEC stacks technology and its adaptation and use for co-SOEC and pressurized operation. It will provide a good international visibility of CEA and will allow to participate in future projects in this field, either national or international, collaborative or bilateral with industrials. It has also gained know-how in the field of testing in particular pressurized testing.

6 The development will serve as a base for a new products being developed in current projects addressing larger power production.
7 All the models developed in the frame of SOPHIA by EPFL and VTT will be used for several forthcoming studies and projects dedicated to SOFC and SOEC.

As a general matter, all the numerical means developed in SOPHIA will be valorized through studies dedicated to the optimization of high temperature fuel cell and electrolyser. They allow to narrow the gap between the laboratory developments and the pre-commercial systems.

Exploitable result: System
1 Pressurised system
2 Pressure difference control

Exploitation plans
1 The lessons learned by HyGear will be applied in future R&D projects in the field of SOFC and SOEC.
2 The methodology will be used in future SOEC- and SOFC systems.

Exploitable result: CSP
1 Solar sreceiver and steam generator capable of being coupled to an electrolysis system
2 Process control system
3 Large scale system development strategy
Exploitation plans
1 A scale-up of the reactor to a larger scale is planned by DLR in a future R&D project
2 Apply the methodology for other processes under development at DLR in other R&D projects
3 Use the knowledge obtained for future projects

List of Websites:
http://www.sophia-project.eu/

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