



Deliverable 2.5:

Report on the stand-alone Electrolyser testing

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| PROJECT & ACRONYM: | Integrated High-Temperature Electrolysis and Methanation for Effective Power to Gas Conversion (HELMETH) |
| TYPE OF PROJECT: | Collaborative project; Co-financed by the European Union's Seventh Framework Programme for the Fuel Cells and Hydrogen Joint Technology Initiative |
| GRANT AGREEMENT NO.: | 621210 |
| TYPE OF DOCUMENT: | DELIVERABLE |
| LEAD BENEFICIARY: | SUNFIRE |
| CONTRIBUTORS: | KIT |
| FILE NAME: | HELMETH_DELIVERABLE_2.5_R2.0 |
| RELEASE: | R.2.0 |
| DATE: | 31/05/2017 |
| ABSTRACT: | Data and calculations for the stand-alone operation of the pressurised electrolyser module |
| CONFIDENTIALITY LEVEL: | |

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DOCUMENT HISTORY:

| <u>Release</u> | <u>Upload date</u> | <u>Status / comments</u> | <u>Author / reviewer</u> | <u>Distribution</u> |
|----------------|--------------------|--------------------------|--------------------------|--------------------------|
| R.0.1 | 13/03/2017 | 1 st Draft | SUNFIRE | Project internal website |
| R.0.2 | 04/04/2017 | 1 st Review | KIT | Project internal website |
| R.0.3 | 20/04/2017 | 2 nd Draft | SUNFIRE | Project internal website |
| R.1.0 | 04/05/2017 | Version 1.0 | KIT | EC participant portal |
| R.1.1 | 15/05/2017 | 3 rd Draft | SUNFIRE | Project internal website |
| R.1.2 | 22/05/2017 | 3 rd Review | KIT | Project internal website |
| R.1.3 | 23/05/2017 | 4 th Draft | SUNFIRE | Project internal website |
| R.2.0 | 31/05/2017 | Version 2.0 | KIT | EC participant portal |

Due date specified in description of work: 31/03/2017 (M36)

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Glossary

| <u>Abbreviation / acronym</u> | <u>Description</u> |
|--------------------------------------|-----------------------------------|
| approx. | Approximately |
| bara | Bar absolute |
| DC | direct current |
| DoW | Description of Work |
| HTE | high temperature electrolysis |
| Max. | maximum |
| OCV | Open circuit voltage |
| PtG | Power-to-Gas |
| resp. | respectively |
| SOEC | Solid oxide electrolyser cell |
| SOFC | Solid oxide fuel cell |
| WP | Work package |
| H ₂ | Hydrogen |
| H ₂ O | Water |
| ISM | Integrated Stack Module (sunfire) |
| ESC | Electrolyte supported cells |
| SC | Steam Conversion |
| BoP | Balance of Plant |
| | |

| <u>Project partner acronyms</u> | |
|--|--|
| KIT | Karlsruhe Institute of Technology |
| POLITO | Politecnico di Torino |
| SUNFIRE | sunfire GmbH |
| ERIC | European Research Institute of Catalysis A.I.S.B.L. |
| EEI | EthosEnergy Italia |
| NTUA | National Technical University of Athens |
| DVGW | DVGW - German Technical and Scientific Association for Gas and Water |

1. Executive summary

This deliverable **D2.5 “Report on the stand-alone electrolyser testing”** is part of the **HELMETH project**, which is devoted to a proof of concept of a highly efficient Power-to-Gas (PtG) technology with methane as a chemical storage and by thermally integrating high temperature electrolysis (SOEC technology) with methanation.

The main objective of this report is to show that the electrolyser module can be operated under pressure and present the experimental characterization of such operation. For this purpose, the system consisting of SOEC stack and hot BoP components have been integrated into a pressure vessel. The system was heated up to operating temperatures $> 700^{\circ}\text{C}$ with electrically heated steam and nitrogen flows. During the testing time of 200 h, variations of the pressure level (3 ... 15 bar), electrolysis power (up to about 8 kW) and steam conversion (50 ... 90%) have been performed.

The results of the pressurized SOEC operation are satisfying, and hence the Milestone 5 “Successful operation of electrolyser module” of the HELMETH project is regarded as fulfilled.

2. Introduction

While a lot of data and information is available on SOC operation from cell level to full scale systems at ambient pressure (fuel cell and electrolysis operation, e.g. (1), (2)), nearly no literature can be found on pressurized operation. Most of the available literature is based on theoretical analysis and simulations (3), experimental data is only available at cell level (4), (5) and very few on stacks (6).

Generally, the influence of pressure on SOEC performance is low. Advantage is seen at system level in an integrated process at higher pressures, e.g. Power-to-Gas with coupled methanation that operates above 10 bar.

Main challenges at this topic are the combination of high temperature (600 ... 900°C) and high pressure (> 10 bar targeted) as well the control of very low differential pressures at a high pressure level.

For low temperature electrolysis (like PEM) pressurized operations seems to be better manageable. The stacks can be built in a way that they withstand high pressures themselves by using a proper thickness of metal plates and clamping force. At high temperatures, the sheet metal plates (interconnects etc.) have very low mechanical strength. When the internal pressure is increased, the plates may inflate and lead to leakages and total stack failure. Similar problems may occur to the hot BoP components.

Therefore, the only way to operate a high temperature system at high pressures seems to be to keep differential pressures over hot parts as low as possible and shift high pressure differences to a relatively cold area. From that point of view, a feasible way is to bring all hot components including the stack inside a pressure vessel. So it is possible to minimize the differential pressure at the hot components and transfer the pressure step to the shell of the pressure vessel. As the dimensioning of the pressure vessel shell thickness depends on pressure and temperature, it is important to monitor or control the shell inner temperature.

3. Experimental

3.1. Test setup

For the tests, sunfire stacks were used. A stack consists of 30 electrolyte supported cells (ESC) with an active area of 127.8 cm² each. The cell has a Ni-GDC (nickel/gadolinium doped ceria oxide) cathode, a 3YSZ (3 mol% yttria stabilized zirconia) electrolyte and a LSCF (lanthanum strontium cobalt ferrite) anode, with several interface layers. As interconnects, sheet metal plates (Crofer 22 APU) with a protective coating to reduce oxidation are used. Gaskets out of glass ensure a very good gas tightness between anode and cathode side.

The environment for the stacks is a sunfire ISM (Integrated stack Module). For the purpose of this test, it contains 3 stacks (so in total 90 cells), temperature sensors, voltage probes, current rods and thermal insulation. The ISM is coupled to a hot BoP unit with a heat exchanger and an electrical heater (3.5 kW) for each, anode and cathode side. Additional, an electrical evaporator is attached to ensure the stand-alone operation, when no external steam is supplied.

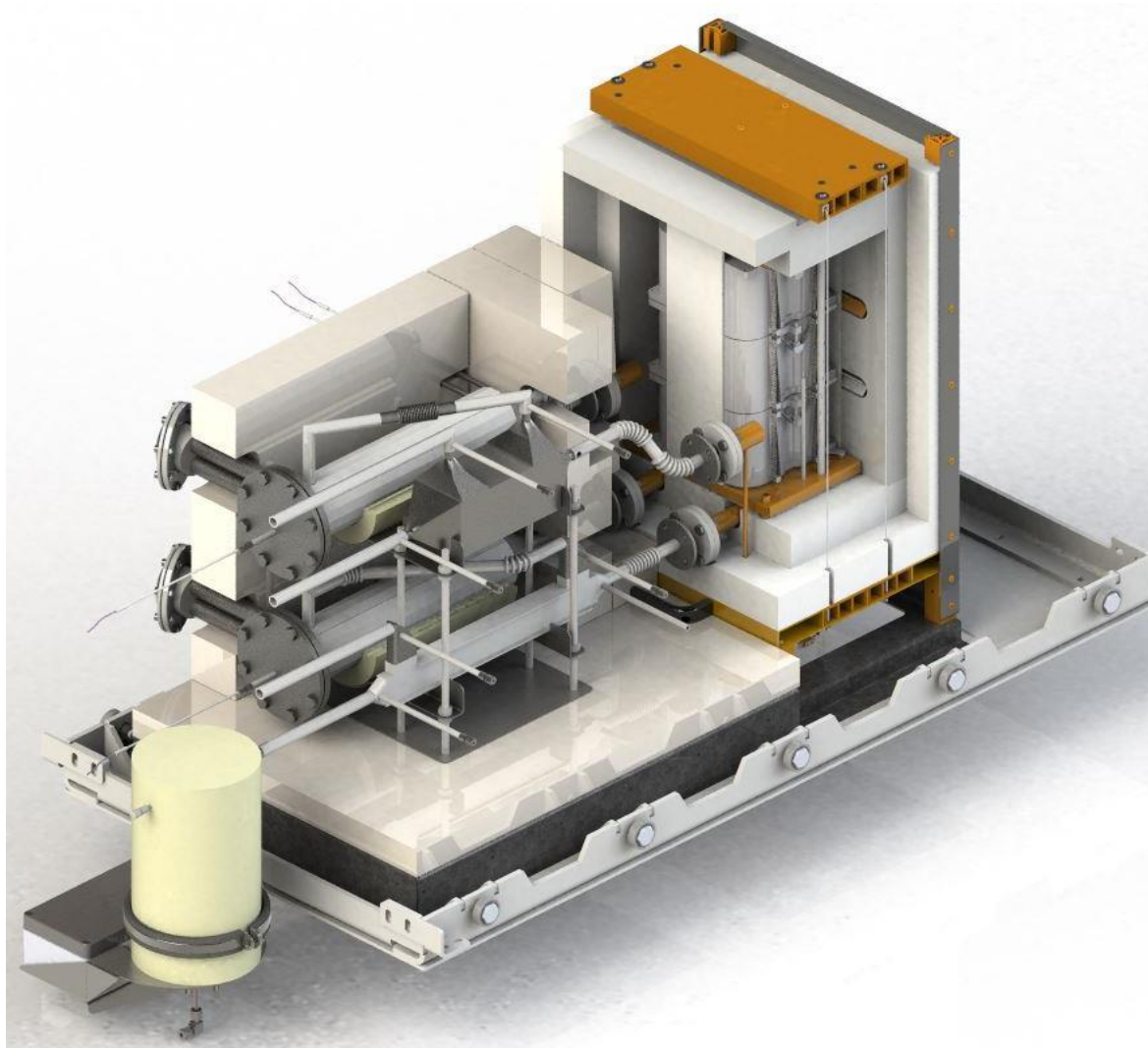


Figure 1: 3D model of (open) hot BOP box and ISM

The evaporator, electrical heaters and heat exchangers are developed by sunfire. The evaporator is a pressure vessel itself, containing heating cartridges. To heat up the gases, direct electrical

heaters with a power of 3.5 kW each are used, containing a heating wire made of Aluchrom. Heat exchangers have a plate-type design and are assembled with laser welding. The design of the Hotbox allows to replace the plate-type heat exchangers with the ones that are developed within the HELMETH project by POLITO, made of Inconel and fabricated in an additive manufacturing process (DMLS – direct metal laser sintering). The assembled unit of ISM and hot BoP box is shown in Figure 2.

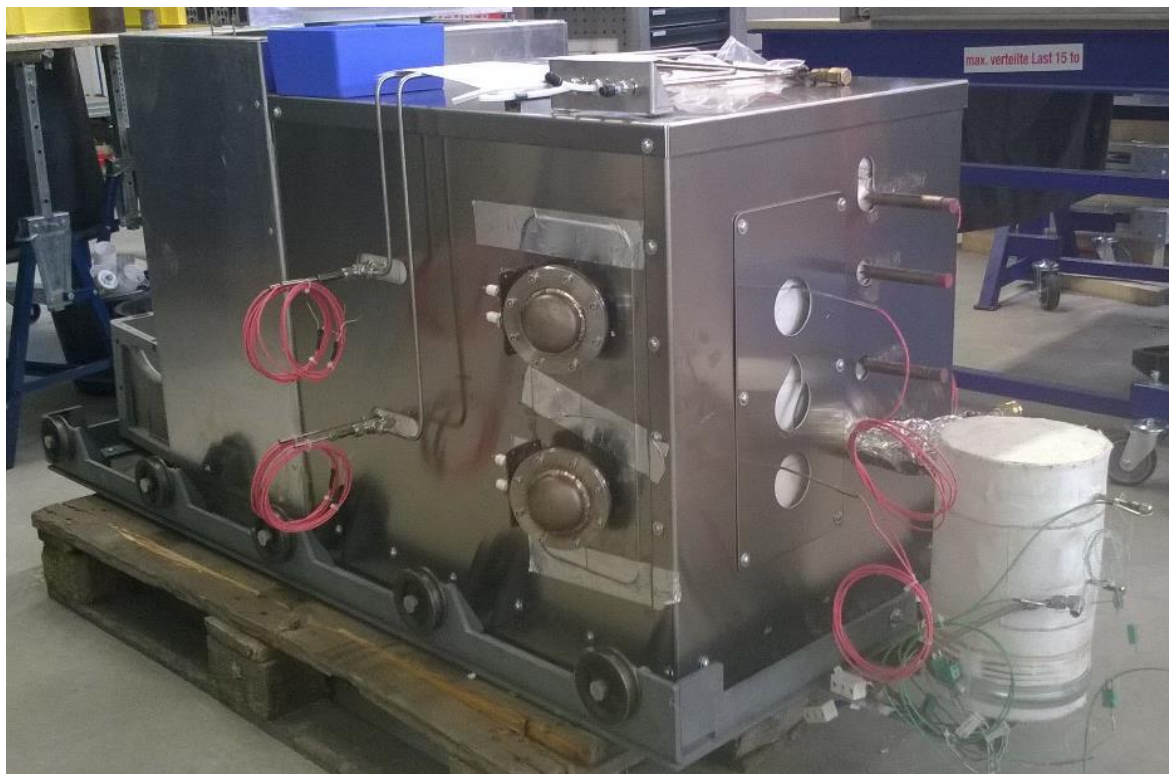


Figure 2: hot BoP box and ISM after production

The test environment was installed in a standard container (20 ft) which is equipped with a full safety system, including hydrogen sensor and smoke detector as well as a ventilation system. To ensure not to exceed the maximum pressures, safety valves and blowouts are connected to the respective gas pipes. The whole system was inspected and approved due to the pressure vessel directive by an approved inspection authority.

Inside the container, the pressure vessel is installed. The pressure vessel is designed for a pressure of up to 30 bar. Additional, all media supply (including filters, pressure regulators, magnetic valves, mass flow controllers), pressure control system (several pneumatic valves), electrical DC power supply for electrolysis and a control cabinet are installed inside the container.

The water supply is done with deionized water from laboratory supply (conductivity < 2 $\mu\text{S}/\text{cm}$), and flow rate is controlled with a membrane pump which is capable to pressurize the water of up to 16 bar. The setup can be seen in Figure 3.

As the final assembling steps, the unit with ISM and hot BoP box is inserted into the pressure vessel (Figure 4), then the pressure vessel is closed, using a graphite seal.



Figure 3: pressure vessel inside container

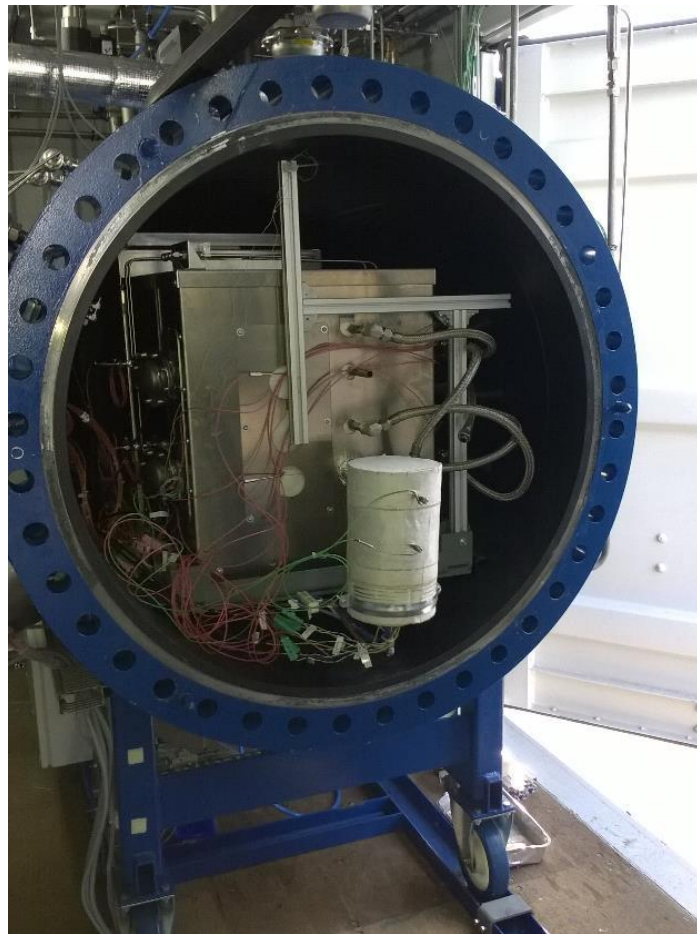


Figure 4: hot BoP box and ISM integrated into pressure vessel

3.2. Test program

Nitrogen (N_2) at the oxygen side of the stack is used to heat the system to temperatures $> 550^\circ\text{C}$. Then, also steam supply (water supply and electrical evaporation) is started at the hydrogen side to enhance heat-up. To ensure a reducing atmosphere at the hydrogen electrode even at OCV conditions, a small amount of hydrogen (H_2 ; 5 Vol.%) is added. The electrical heaters for N_2 and steam are set to an outlet temperature of 820°C . Parallel to the increasing temperatures the pressure level is elevated to a minimum of 3 bar.

Pressure control is aiming to minimize the differential pressure between hydrogen side and oxygen side of the stack. Independent of the absolute pressure, the differential pressure should be kept < 50 mbar.

At the beginning, the steam supply was fixed to 2 kg/h. The current density of electrolysis was increased stepwise to adjust the steam conversion from 50% to 90%.

For the operation at nominal power of the ISM, SC = 70% was chosen. Current density was set to 0.52 A/cm^2 with a steam flowrate of 2.88 kg/h.

Next, the pressure level was increased in 1 bar steps up to 15 bar. Focus at this part of the test was the pressure dependency of the heat losses.

4. Results

4.1. Total test run

The pressurized HTE system has already operated for more than 200 hours and the test is still continuing. One focus was the evaluation of the control of the pressure difference between hydrogen side and oxygen side of the stack at different conditions.

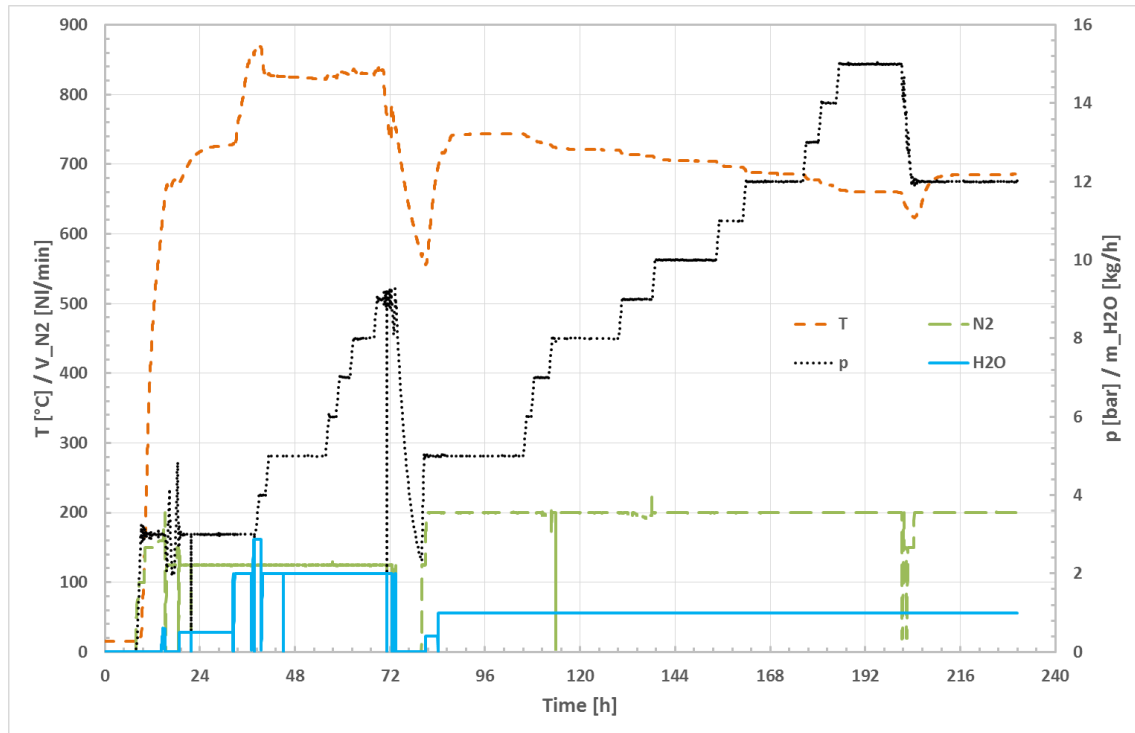


Figure 5: complete test run

The differential pressure could be controlled in a way that it did not exceed ± 100 mbar in all operation points, even in the transient states. From that point of view, a safe operation for the SOE stack was possible. In addition, the differential pressure could be set to any value in that range, meaning that dependent on the specific requirements low overpressure on hydrogen side or oxygen side could be fixed.

4.2. Variation of steam conversion

In order to achieve high system efficiencies it is required to operate the stack with high steam conversion (SC). Results from cell or stack level are often obtained at SC = 50%. To be competitive at system level a SC > 80% is targeted. At the test run, pressure (3 bar), nitrogen flow rate (125 Nl/min) and water flow rate (2 kg/h) have been fixed. Then, current was increased stepwise from 0 A to 60 A, corresponding to a current density of $j = 0.47$ A/cm².

$$SC = \frac{N \cdot I \cdot 60 \cdot V_m}{2 \cdot F \cdot \dot{V}_{n,steam}}$$

N: number of cell (here: 90) I: current [A] V_m : molar volume (22,413 l/mol)
 F: Faraday constant (96485,3 C/mol) $\dot{V}_{n,steam}$: steam flow rate (l/min)

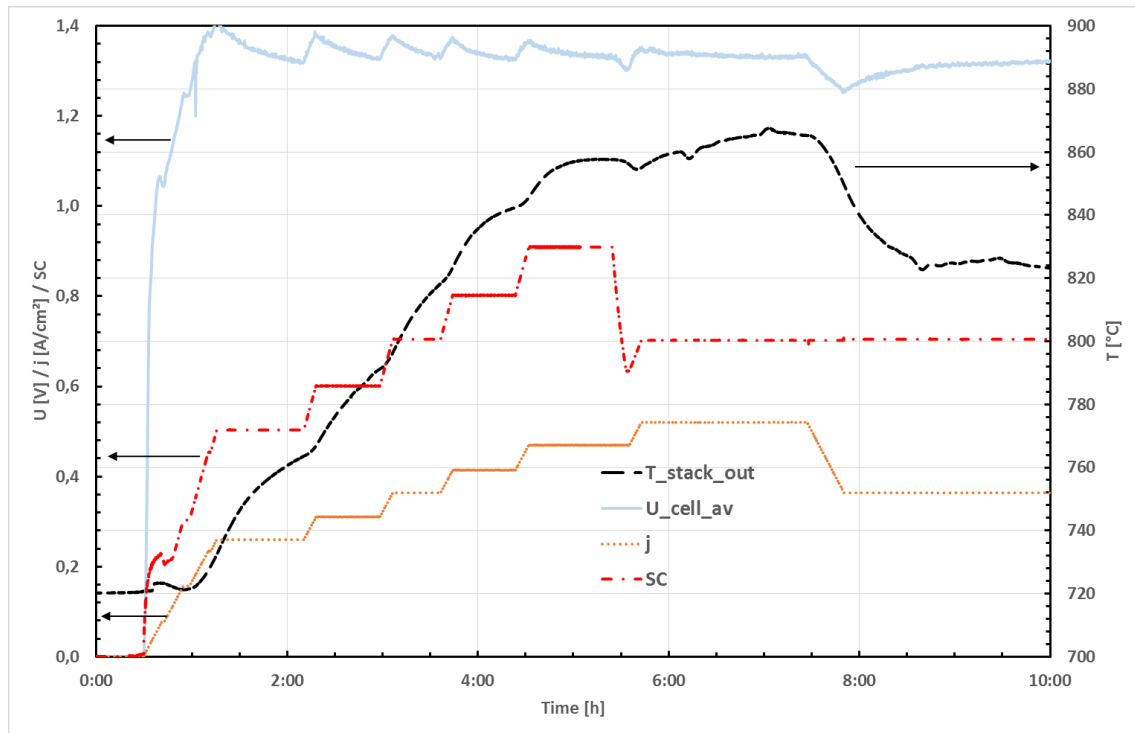


Figure 6: Variation of steam conversion at constant gas flow rates

The stack temperature rises within this test from 730°C at OCV conditions to a maximum of 865°C at full load (Figure 6). This effect is caused by the internal heat release inside the stack due to its internal ohmic resistance. Therefore, a part of the electrical energy supplied to the stack for electrolysis is converted into heat. With rising temperature at higher currents, the internal resistance of the stack is decreasing due to the temperature dependence of the resistance (resp. conductivity) of the ceramic electrolyte and electrodes. This effect counterbalances the increase of the heat release due to Ohmic losses, so that the ratio between internal heat release and load remains almost constant and thus also the average voltage per cell is stabilized at about $U_{cell} \approx 1.3$ V (equals 117 V for 90 cell stack) independent of the applied load. The achieved operational voltage is almost equal to the thermoneutral voltage. At this point, the total energy demand of endothermal steam electrolysis is equal to the supplied electrical energy covering both, the energy for reversible electrochemical conversion and the additional heat demand for the overall reaction.

4.3. Variation of pressure

As reference load point, $I = 46.5$ A and $SC = 70\%$ was chosen. Starting from 5 bar, the pressure level was increased in steps of 1 bar.

Figure 7 shows the development of the stack voltage and temperature. The pressure was increased before total equilibrium of temperature and voltage was reached. It was observed that with increase of pressure the stack temperature rises initially before levelling down again. Anyway, the data can lead to the assumption that there is a slightly higher temperature with increased pressure.

So far, there is no real explanation for this behavior. Possibly it is just an effect of measurement accuracy. With increased pressure, the volume flow rate is getting lower. The temperature sensors

(thermocouples) will be influenced more by radiation than by gas flow temperature. As a second approach of an explanation, a complex thermal management is considered. With increasing

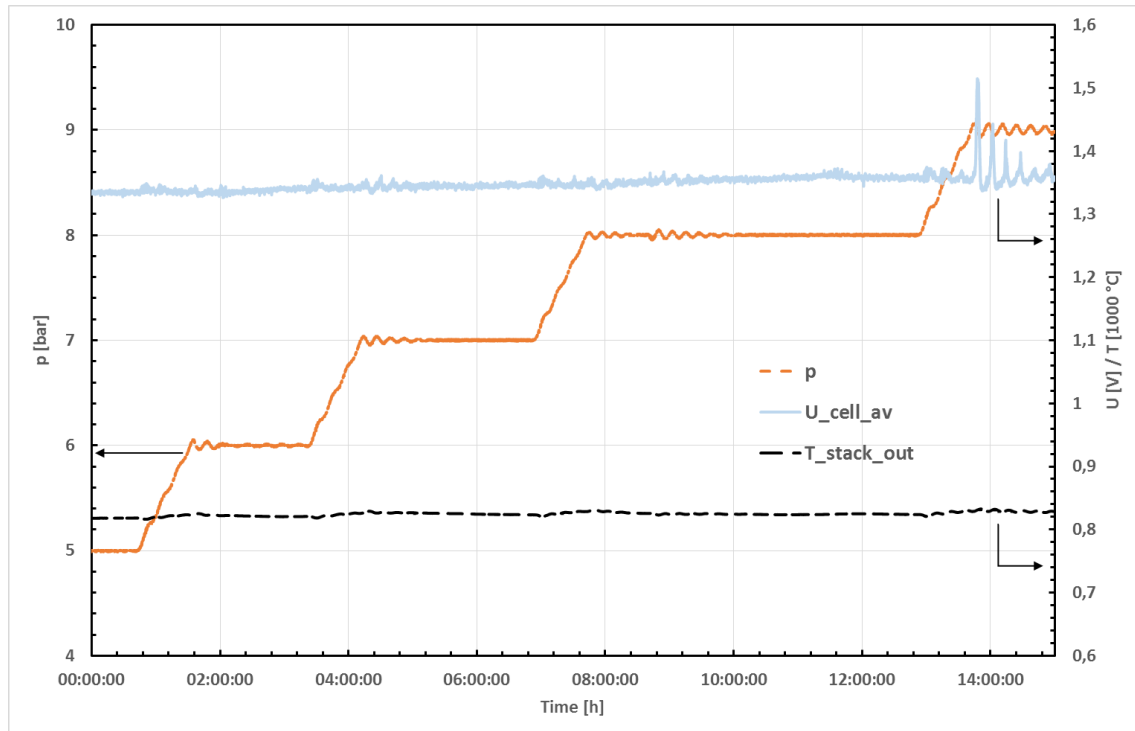


Figure 7: increasing pressure at constant current and flow rates

pressure, the measured temperatures get lower (see 4.4). Therefore, the stack needs to produce more heat itself to compensate the heat losses.

4.4. Heat losses with increased pressure

It is known from previous tests that the microporous insulation material used in hot BoP box and ISM as thermal insulation shows a decreased performance with increasing pressure. Nevertheless, based on the performance at ambient pressure, no proper alternative was found, especially as performance data over pressure is hardly available at the suppliers. Discussion with the manufacturer of the used insulation material is ongoing. It is assumed that due to the open porous structure heat conductivity is enhanced by the more dense gas at elevated pressure. Moreover, also natural convection heat transfer to the insulated walls of the vessel is increased at elevated pressure.

In order to assess the heat losses with increased pressure an experiment was performed under preheating operation conditions without electrolysis mode. The pressure level was increased from 5 bar to 15 bar, while the gas mass flow rates were kept constant (steam: 1 kg/h; N₂: 200 NI/min), as well as the outlet temperatures of the electrical gas heaters (820°C). The measurement point of steam and N₂ gas inlet temperature is in the corresponding feed pipes to the ISM. These thermocouples are not influenced by heat radiation of the stack. Stack temperature itself is measured in layer 45 (of 90) directly at the gas outlet of the MEA. This thermocouple is influenced by the heat radiation of the neighbouring interconnects.

Figure 8 shows the test results. The N₂ inlet temperature decreases from 795°C to 745°C, while steam temperature drops from 680°C to 490°C. This is likely related to significantly lower volume flow rate and therefore lower gas velocity. The stack core temperature decreases from 745°C to 660°C. This proves that the stack is mainly heated by the nitrogen flow and the low steam temperature has only minor influence.

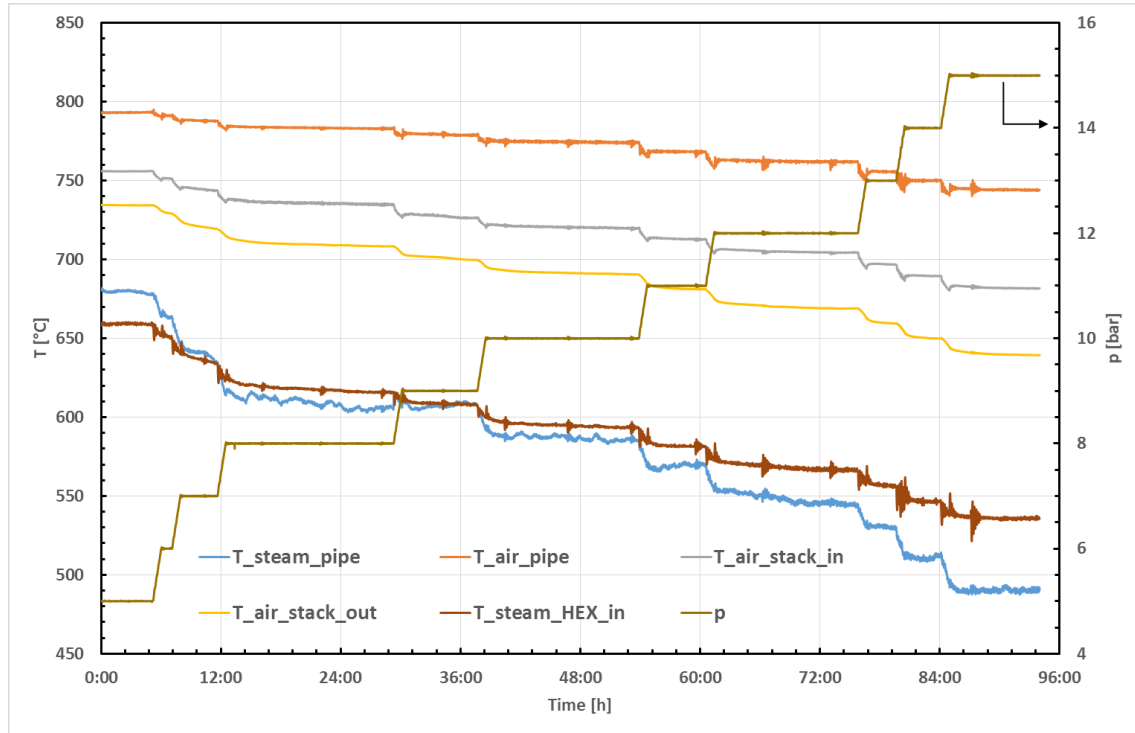


Figure 8: dependency of temperatures from pressure at constant gas flow rates

4.5. Efficiency

In terms of possible business models, the efficiency of an electrolysis system is of high importance. The reason is that the overwhelming part of the total operation costs (OPEX) comes from the required electrical energy. To justify higher investment costs (CAPEX) for a high temperature electrolyser, it has to have higher efficiencies than the competing technologies.

The efficiency can be calculated at different levels. First is the stack efficiency itself, only taken into account the produced amount of hydrogen and electrical DC power input:

$$\eta_{Stack} = \frac{\dot{V}_{H_2-Output} \cdot HHV_{H_2}}{P_{el_DC_Stack}} = \frac{c}{U_{Stack}}$$

Equation 1

The HHV of Hydrogen is 3.54 kWh/Nm³. Next step is to consider the heat losses. The assumption is that the heat losses are balanced by the two electrical heaters. As the AC side of the system is not measured, the power of the electrical heaters (as well as the evaporator) are considered as the nominal AC power multiplied with the control signal (PWM – pulse width modulation):

$$\eta_{Heat_Loss} = \frac{\dot{V}_{H_2-Output} \cdot HHV_{H_2}}{P_{el_DC_Stack} + P_{el_AC_Gas_Heaters}}$$

Equation 2

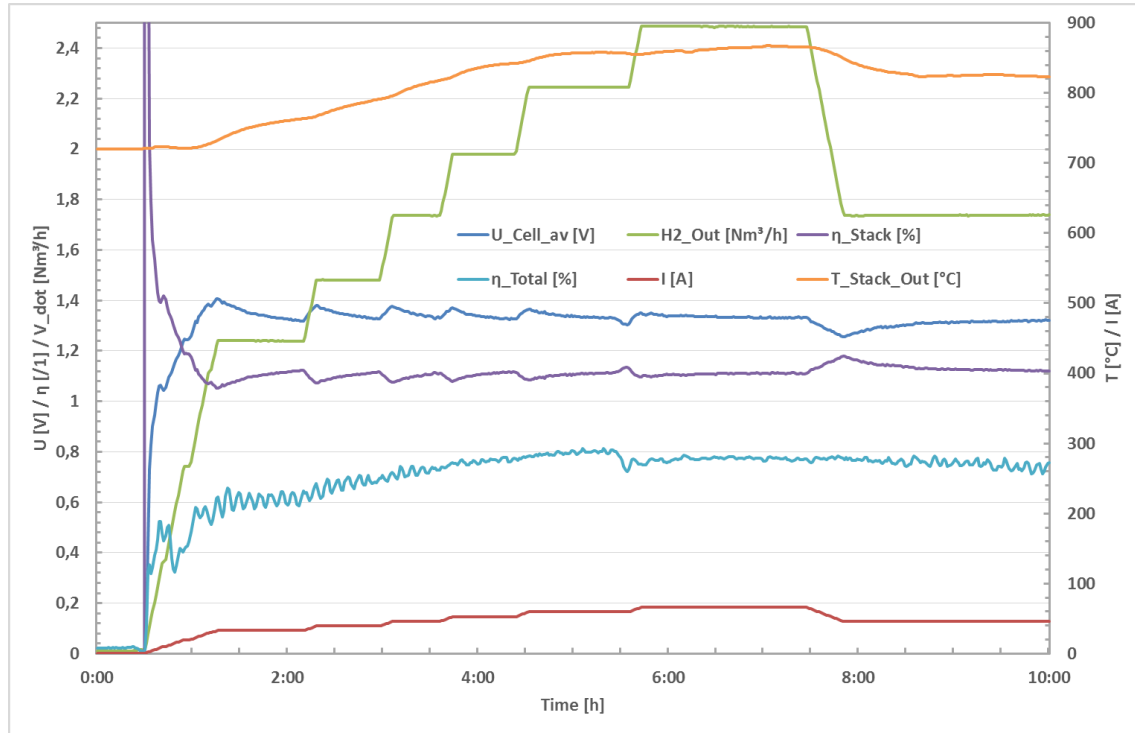


Figure 9: Efficiency of Stack itself and with compensation of heat losses and evaporation

To complete the picture, the electrical evaporation has to be taken into account for this specific system:

$$\eta_{Total} = \frac{\dot{V}_{H_2-Output} \cdot HHV_{H_2}}{P_{el_DC_Stack} + P_{el_AC_Gas_Heaters} + P_{el_AC_Evaporator}}$$

Equation 3

Figure 9 shows the average stack temperature, the total current and the corresponding hydrogen production, average cell voltage and the efficiencies of the stack and the system. The belonging steam conversions are shown in Figure 6.

The stack efficiency is independent of the current and the steam conversion. The reason is according to Equation 1 that η_{Stack} is only indirectly proportional to the stack voltage, while current and hydrogen production are related by a constant factor and HHV is also constant. In case the stack is operated close to the thermoneutral voltage (approx. 1.25 to 1.3 V / cell), the electrical efficiency of the stack is about 110% (peak at the beginning due to measurement inaccuracy at low current). The reason for electrical efficiencies above 100% is that the required energy for the electrolysis is partly resulting from the thermal energy of the steam.

When the electrical heaters and the electrical evaporator are taken into account the picture is changing. The efficiency η_{Total} drops to values about 75 ... 80%. This is not surprising, as the electrical evaporation of water is energy intensive, because the evaporation enthalpy has to be overcome. For that reason, it is highly recommended that high temperature electrolyser systems are coupled to existing steam supplies. These are available in many industrial facilities. Industrial used steam often has a remaining temperature of > 150°C, which is sufficient for atmospheric HTE systems.

To operate at higher pressures, the excess steam requires temperatures of above 200°C, which corresponds to a pressure of about 15 bar. These are pressure levels that can be found in PtL and PtG processes like methanation. The coupling of HTE and methanation as in HELMETH shows the potential of a fully integrated process (integrated at temperature and pressure side).

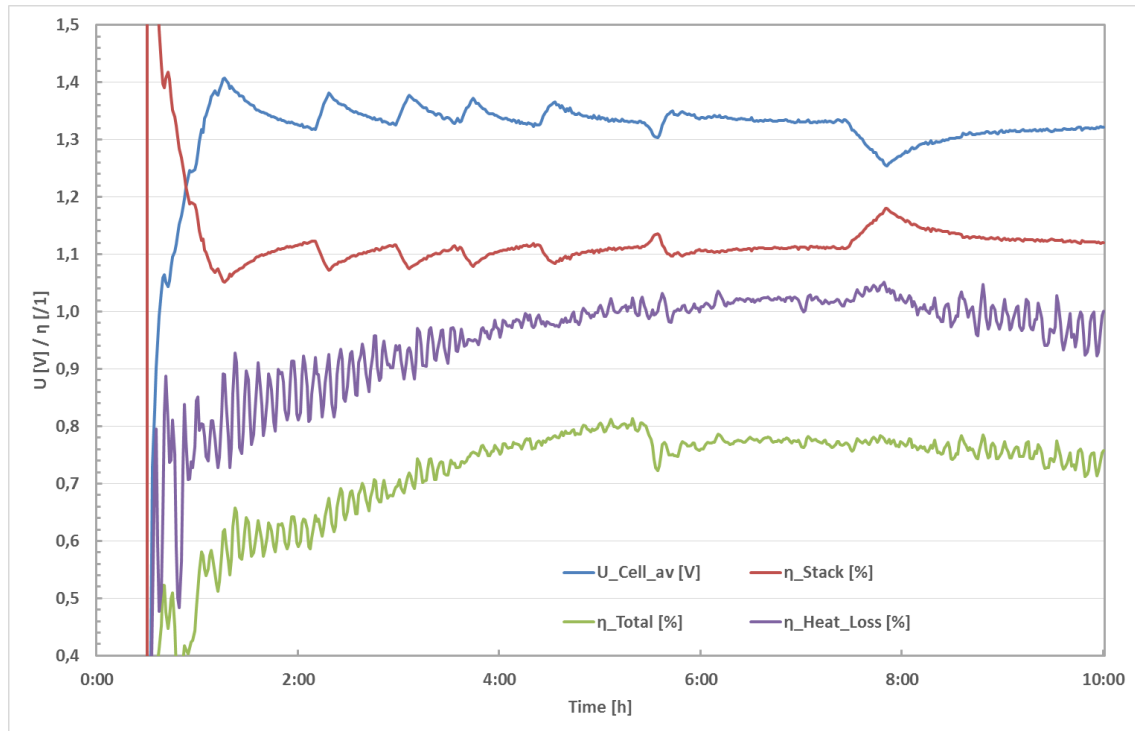


Figure 10: Efficiency shares of heat losses and evaporation

To show the influence in case steam can be used instead of water, in Figure 10 the share of the efficiency only with the electrical heaters and stack is shown.

With increasing hydrogen production (that means increasing current and increasing stack operation temperature) the efficiency of stack and heaters ($\eta_{\text{Heat_Loss}}$) rises to above 95% close to 105%. As the gas flow rates and the heater output temperatures are kept constant, a higher stack temperature leads to higher heat transfer in the heat exchangers and therefore less requirement of electrical heating. In a real system, it is likely that also in part load conditions the efficiencies will be high, as flow rates and, if required, heater temperatures will be adjusted to the operation point.

In addition, this means the electrical evaporation during this test “costs” about 20% of electrical efficiency. Besides the evaporation itself, this value contains the heating of water from room temperature to its evaporation temperature and an overheating of the steam to about 300°C.

Summary and conclusion

It has been shown that the design, manufacturing and operation of a pressurized high temperature electrolysis has been successful at sunfire. The differential pressure between hydrogen side and oxygen side can be controlled in a wide operation range to values well below 100 mbar and in most cases even below 50 mbar. The absolute pressure can be varied up to 15 bar. The SOC stacks from sunfire are able to be operated with a steam conversion of up to 90% which is important to gain high system efficiencies.

In addition to the stack performance data, a clear relation between pressure and heat losses was observed. Two major aspects are responsible for the influence. First, with increasing pressure the flow velocities get lower. Although the mass flows stay constant, in a non-ideal system lower flow velocities increase the relative heat losses, e.g. heat transfer inside the plate-type heat exchangers is influenced negatively. Second, the used microporous insulation which is a very good thermal insulator at ambient pressure shows a decrease in performance with increasing pressures. The effect is not fully understood yet, discussions with the supplier are ongoing.

Regarding the efficiency, the performance of the stack corresponds to the expectations with values about 110% and no dependence on the steam conversion. The heat losses (heat transfer efficiency of the heat exchangers and heat losses to the environment of the hotbox) are in the range of < 1 kW electrical heater power, which corresponds to a decrease in electrical efficiency of about 10% to 100%.

The main decrease in efficiency to overall about 75...80% of the SOEC module is resulting from the energy input to the electrical evaporation unit. This is a major reason while one is aiming to use steam produced by excess heat elsewhere, as in the planned coupling with the methanation unit within HELMETH.

Based on these results, Milestone 5 of the HELMETH project "Successful operation of electrolyser module" is reached.

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