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Solar hydrogen by high-temperature electrolysis: Flowsheeting and experimental analysis of a tube-type receiver concept for superheated steam production

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Abstract

High-temperature electrolysis (HTE) offers the potential of considerably higher efficiency than conventional alkaline electrolysis when producing hydrogen from water by solar energy. The production rate of the alkaline electrolyzer process is limited since the whole energy demand is covered by electricity. By contrast, in HTE part of the energy can be introduced as high temperature heat from concentrated solar power (CSP) leading to a significantly higher process efficiency. In the internal project SoHTEk a solar tube-type receiver to superheat steam to 700 °C for HTE was developed. The receiver was operated in DLR's solar simulator with up to 5 kg/h of steam reaching an outlet temperature of about 700 °C at a thermal efficiency of 40 % and a solar power of about 4 kW. In addition, a flowsheeting analysis was carried out of a HTE process with direct steam generation on a solar tower. Simulation of the process at steady-state yielded a thermal-to-hydrogen efficiency of 26 %. A subsequent sensitivity analysis indicated the potential to reach efficiencies of 38 % and more.

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1. Introduction

Water electrolysis is considered as a promising pathway for a sustainable hydrogen production. There are two main water electrolysis processes depending on the operation temperature: alkaline electrolysis and high

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temperature steam electrolysis (HTSE). The total energy demand of alkaline electrolysis is supplied as electricity. By contrast, a part of the total energy is introduced as high temperature heat in the HTSE process. As compared to conventional alkaline electrolysis, HTSE achieves higher efficiency. High temperature heat and electricity requirements of the electrolyzer can be provided by concentration of solar radiation in solar towers. Conceivable heat transfer fluids are e.g. water/steam and molten salt. A solar tower plant with water as working fluid, also known as direct steam generation in central receiver system will be investigated in terms of electricity and heat generation in order to run the HTSE process. Operating the process with direct steam from a CSP tower has the benefit of using a heat transfer fluid well suited to generate electricity in a Rankin cycle as well as feed steam for the HTSE. Moreover, this process scheme permits the use of steam as sweep gas for the anode. Following analysis is carried out in addition to the EU project ADEL [1], which investigates the integration of suitable heat and power sources, e.g. CSP technologies, in the HTSE process. The designs of the HTSE process and its requirements have been already defined in the mentioned project. Evaporation and superheating of water for the power block and HTSE unit take place in a solar tower receiver. The superheating section of the receiver as a key component of the process was designed by DLR within the internal project SoHTEk. This tube-type test receiver was developed for operation in DLR's high flux solar simulator in Cologne. Moreover, a flowsheet of a plant in MW-range was elaborated and the process was simulated with the commercial software tool Aspen Plus. This paper will provide an overview of the solar high temperature electrolysis with focus on the design of the solar receiver and the coupling of high temperature steam electrolysis to direct steam generation in a central receiver system.

Nomenclature

A	surface area
α	heat transfer coefficient
d	diameter
ε	emissivity
φ	view factor
H	enthalpy
l	characteristic length
λ	heat conductivity
$\dot{m}$	mass flow
$\dot{Q}$	heat flow
Nu	Nusselt-number
η	efficiency
P	Power
Pr	Prandtl-number
Re	Reynolds-number
σ	Boltzmann-constant
W	Work/Energy

2. Thermodynamics of solar high-temperature electrolysis

The splitting of water can be achieved through the high temperature electrolysis of steam (HTSE), which uses a combination of electrical energy and heat. The chemical reactions, which take place in the electrolyzer, are given as follows:

- On the cathode side: $H_2O + 2e^- \longrightarrow H_2 + O^{2-}$
- On the anode side: $O^{2-} \longrightarrow \frac{1}{2}O_2 + 2e^-$

Both reactions give the overall water splitting reaction:

$$H_2O \longrightarrow H_2 + \frac{1}{2}O_2 \qquad (1)$$

Water is dissociated at the cathode into hydrogen and oxygen ions by electrical current and heat. The oxygen ions pass through the electrolyte because of the electrical field between the cathode and the anode, where the oxygen ions will be oxidized in order to form oxygen molecules.

Depending on the cell voltage, there are three different operation modes for the electrolyzer:

- *Thermoneutral mode:* in this operation mode, the electricity supplied to the electrolyzer is completely used for the electrolysis reaction. As a result, the outlet gases (H_2/H_2O and O_2) have the same temperature as the inlet gas (H_2/H_2O).
- *Exothermal mode:* in this operation mode, the cell voltage is higher as the voltage needed to split the water molecule. Consequently, a part of the energy will be converted into heat, which will be absorbed by the outlet gases.
- *Endothermic mode:* the outlet gases have a lower temperature than the temperature of the inlet gases since the heat production in the cell is lower than the heat absorbed by the reaction.

In this study, the high temperature steam electrolyzer will be operated in the thermoneutral mode based on the electrolyzer model validated in the EU project ADEL [1].

3. Solar receiver for superheated steam production

3.1. Development of a solar tube-type receiver

To provide the electrolysis with superheated steam at a temperature level of 600 to 700 °C a solar tube-type receiver with a cylindrical configuration has been developed as depicted in figure 1 [5]. Saturated steam of 100 °C enters the absorber pipes from behind and is superheated to 700 °C by concentrated solar radiation.

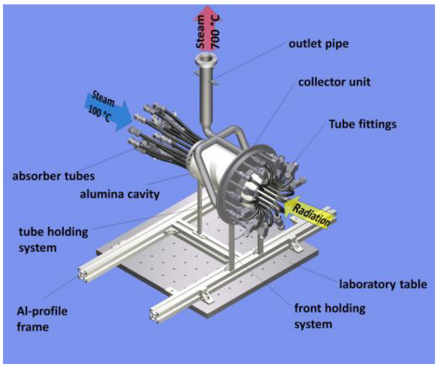

Fig. 1. CAD-model of solar receiver [5].

The receiver's layout was carried out applying the equation solver EES® and the ray tracing tool OptiCAD®. The first step was to select the geometry. The aperture diameter is fixed by the nominal diameter of DLR's solar simulator focal point of 90 mm [6]. Steel tubes with 10 mm outer diameter were chosen because of their good availability off the shelf. Therefore, the amount of tubes was set to 20 in order to shape the required aperture. The remaining degrees of freedom for the geometry (i.e. length of tubes) were calculated by applying an integral heat balance of the system. To do this a tube wall temperature of 900 °C was assumed. On the steam side the heat flux is given by the energy amount for superheating steam from 100 °C up to 700 °C.

$$\dot{Q}_{fluid} = \dot{m}_{fluid}\left(H_{fluid,T_{700}} - H_{fluid,T_{100}}\right) \qquad (2)$$

This equation needs to be equated to the heat income and the heat losses:

$$\dot{Q}_{fluid} = \dot{Q}_{In} - \dot{Q}_{conv} - \dot{Q}_{rad} \tag{3}$$

$\dot{Q}_{In}$ is the incoming radiation provided by the solar simulator, $\dot{Q}_{conv}$ are the convectional heat losses, both natural and forced and $\dot{Q}_{rad}$ are the losses by reradiation. The incoming radiation is given by the solar simulator's maximum output of around 15 kW [6] which can be adjusted from 0-100 % by a shutter system. For geometry layout only forced convection $\dot{Q}_{Fconv}$ was considered neglecting natural convection on the outside of the tubes.

$$\dot{Q}_{Fconv} = A_{tube} \cdot \alpha_{FC} \cdot \Delta T_m \tag{4}$$

A_{tube} is the heat transfer area of all tubes, ΔT_m is the average logarithm temperature of the fluid and α_{FC} is the heat transfer coefficient for forced convection. This α_{FC} was calculated with a Nusselt-correlation found in the VDI-heat atlas [7]:

$$Nu_m = \alpha_{FC} \cdot \frac{l_{cha}}{\lambda} \tag{5}$$

$$Nu_m = \left(3{,}66^3 + 0{,}7^3 + \left(1{,}615 \cdot \left(\mathrm{Re} \cdot \mathrm{Pr} \cdot \frac{d_i}{l_{tube}} \right)^{1/3} - 0{,}7 \right)^3 + \left(\frac{2}{1 + 22 \cdot \mathrm{Pr}} \right)^{1/6} \cdot \left(\mathrm{Re} \cdot \mathrm{Pr} \cdot \frac{d_i}{l_{tube}} \right)^{1/2} \right)^{1/6} \tag{6}$$

With l_{cha} the characteristic length, d_i the tube's inner diameter and l_{tube} the total tube's length. To determine the radiation heat losses, the view factors had to be calculated. For the purpose the cylindrical receiver was approximated by three areas: 1) inlet aperture, 2) mantle area surrounded by tubes and 3) the receiver's closed backside. The main equation was the relation between area 1 and 3 [8]:

$$\varphi_{1-3} = \tfrac{1}{2} \left[X - \left(\sqrt{X^2 - 4 \cdot \left| \frac{R_3}{R_1} \right|^2} \right) \right] \tag{7}$$

X and R_1, R_3 are geometric factors. All other view factors could be derived with the law of reciprocity and/or the law of summation [7]. The view factors are used for the radiation equation:

$$\dot{Q}_{rad,i-j} = \sigma \cdot \varepsilon^2 \cdot A_i \cdot \left(\frac{\varphi_{i-j}}{1 - \left(1 - \varepsilon^2\right) \cdot \varphi_{i-j}{}^2} \right) \cdot \left(T_{W,i}{}^4 - T_{W,j}{}^4 \right) \tag{8}$$

$\dot{Q}_{rad,i-j}$ is the radiation exchange between a segment i onto a segment j with a temperature of $T_{w,i}$ and $T_{w,j}$, respectively. The energy balance for a steam outlet temperature of 700 °C was solved in a parametric study resulting in a required minimum tube length of 240 mm. To have some buffer in the heating system, the length was set to 270 mm.

Subsequently, a differential calculation was executed dividing the absorber tubes into 10 segments in axial direction. View factors where formulated for each ring element to account for radiative heat exchange while the forced convection was calculated with the outlet temperature of the segment i forming the inlet temperature of segment i+1 [9]. To determine the solar simulator's heat flux on the segments and locate hot spots, a ray tracing analysis was carried out. The incoming power on the ring elements is depicted in figure 2 showing the distribution of incoming radiation with respect the offset of the focus. A receiver position of 60 mm behind the focus was chosen (red columns) to achieve higher incoming power in the front than in the back and no hot spots in the mid. This is to maximize the heat transfer to the steam flowing from the back to the front.

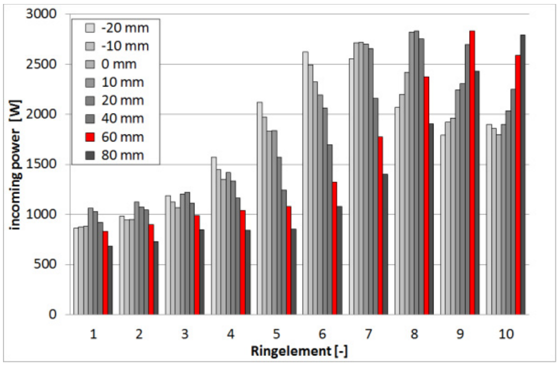

Fig. 2 focus variation (ring 1 at the backside and ring 10 at the aperture)

3.2. Experimental analysis of receiver in high-flux solar simulator

The receiver was successfully started up and tested in the solar simulator of DLR in Cologne (figure 3). The first campaign showed the operability of the set-up with nitrogen while systematic test runs with steam were conducted in the following campaigns. In the third campaign a water cooled cover was installed to reduce spillage of the solar radiation and heating of the housing. Otherwise this heat input leads to a non-regular increase of efficiency.

During the experiments the mass flow of steam and the incoming heat flux were the varying parameters. Figure 4 shows the temperature distribution of one exemplary experiment with the inlet temperature on the primary, left axis and the outlet temperature on the secondary axis. As seen, the aspired outlet temperature of 700 °C can be reached (lower red dotted line). Moreover, the different heating phases are depicted: to heat the receiver to 600 °C around two hours are needed (under N_2 atmosphere). When steam feeding is started and nitrogen is turned off the temperature temporary rises to a peak of about 850 °C. This is due to the missing mass flow, until the steam reaches the thermocouples in the absorber tubes; afterwards it drops again to a reliable value. Waiting for the stationary state with a mass flow 3 kg/h of steam takes around 1.5 hours with a temperature of around 700 °C and 90 % shutter opening. At 4 kg/h of steam the shutter was opened to 100 %, leading to an outlet temperature around 720 °C at the stationary condition. Feeding 5 kg/h of steam, the temperature drops again to around 700 °C. Cooling the receiver, was carried out with N_2 while turning off all lamps. The outlet temperature is higher than the expected 100 °C, because of a higher evaporator temperature, around 190 °C to avoid condensation in the feeding system. Another reason for this is the high conduction of the absorber pipes. They are conducting the heat beyond the irradiated part and preheat the steam. The receiver reaches a thermal efficiency of 40 % at 5 kg/h of steam and a solar power input of around 4 kW.

Fig. 3 irradiated receiver

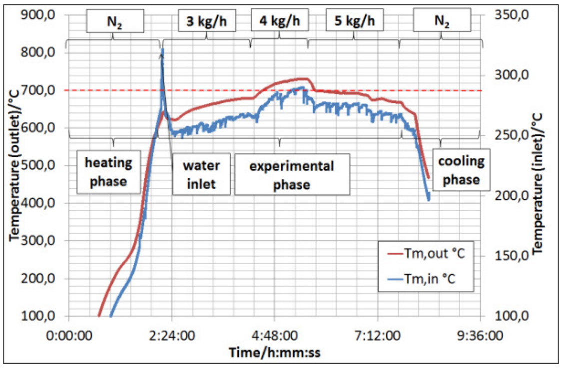

Fig. 4 experimental analysis

4. Process for solar high-temperature electrolysis

4.1. Process design

Central Receiver Systems (CRS) are composed of sun-tracking mirrors, called heliostats, which concentrate solar radiation onto a receiver placed on the top of a tower. In the solar receiver, the solar radiation is absorbed by a heat transfer fluid (HTF) that flows through the tubes of the receiver. The process design of the plant depends on the heat transfer fluid. The heat transfer fluid of the plant analyzed in this study is water. This technology has the advantage of reducing the investment cost since a steam generator is not required. As compared to the other mentioned heat transfer fluid, the steam temperature is not limited by the degradation which allows the use of higher performance Rankine cycles at higher temperature and operating pressure.

In the last ten years, several efforts have been dedicated in order to design new concepts which allow the operation of the plant at higher temperature and pressure. The main characteristics of current Direct Steam Generation –Cental Receiver Systems plants (DSG-CRS) are presented below:

- PS-10 and PS-20 plants with an electricity output of successively 10 and 20 MWe are operated with saturated steam at 250°C and 40 bar. The performance of the PS 10 plant under nominal conditions is about 21.2% [10].
- Sierra Sun Tower pilot plant operated by e-Solar since 2009 generates steam at 440°C and 60 bar. The electricity output of the plant is about 5MWe [11].
- BrightSource is operating a power plant since 2008, where steam is generated at 530°C and 130 bar. Currently, the industrial company is developing a solar power complex using steam as heat transfer fluid at 560°C and 160 bar [12].

In this study, the integration of the HTSE unit in a DSG-CRS plant will be analyzed. The feed water enters to the Rankine cycle with environmental conditions (25°C, 1.013 bar). The conditions of the superheated steam are 530°C and 160 bar. Steam will also act as sweep gas. Using steam as sweep gas in comparison with other fluids has many advantages: steam does not contain oxygen and as a result, the Nernst potential will be reduced and the efficiency of the electrolysis process will increase [13]. Additionally, the outlet stream contains oxygen and steam, which can be easily separated by condensation and oxygen, can be considered as a by-product. The sweeping steam is separately produced by a part of the steam generated in the solar receiver. This will be extracted after the high pressure turbine. The steam turbine consists of two high pressure stages, a medium stage and three low pressure stages. Additionally, a part of the steam generated in the receiver is used to reheat the steam after the second high pressure stage in order to raise the temperature and accordingly the efficiency of the Rankine cycle. The water introduced to the solar receive is preheated in five heat exchangers, which use the receiver steam as a heat transfer fluid. This will be further explained in the next section. After the preheating, the water is evaporated and superheated in the solar receiver.

The HTSE steam is preheated and evaporated by the receiver steam as already mentioned. Then, it is further heated in two heat exchangers. These are recuperative heat exchangers that use heat from the exhaust sweeping gas and the product gas. An electrical heated may be needed in order to rich the operating temperature of the electrolyzer. Finally, the hydrogen produced in the HTSE is compressed for storage after condensation of the remaining water. The next table shows the specifications of the HTSE unit. The specifications are based on the assumptions and results of the EU- founded project ADEL [1].

Table 1. Main specifications of the HTSE unit

Operation Mode	Thermoneutral
Flow rate of HTSE feed water	270.01 kmol/h
Sweep gas/cathode stream ratio	1:1
Steam conversion	60%
Operating pressure	1.5 bar
Pressure drop in the stack	0.5 bar

Inlet/outlet cathode temperature	700°C
Inlet/outlet anode temperature	700°C
H_2% at cathode inlet	10%

Pumps are defined with the mechanical efficiency while the isentropic efficiency is used for the specification of the compressors. It is assumed that the mechanical efficiency of the pumps is 90%. An isentropic efficiency of 85% was selected for the design of the high and medium pressure steam turbines. The isentropic efficiency of the low pressure steam turbines is 75%. A hot/cold temperature approach of 20 K was selected for the simulation of the preheaters. Furthermore, a pressure drop of 3 bar was assumed for the solar receiver, which was simulated as two heat exchangers (evaporation and superheating).

4.2. Steady-state simulation of the process

The steady state simulation of the process has been carried out with the commercial simulation tool Aspen Plus, which is a common tool for the simulation of chemical processes. The flow sheet of the overall process was elaborated with the tool Microsoft Visio. The following figure shows the flow sheet of the DSG plant coupled to the HTSE unit. In the next section, the overall process will be described.

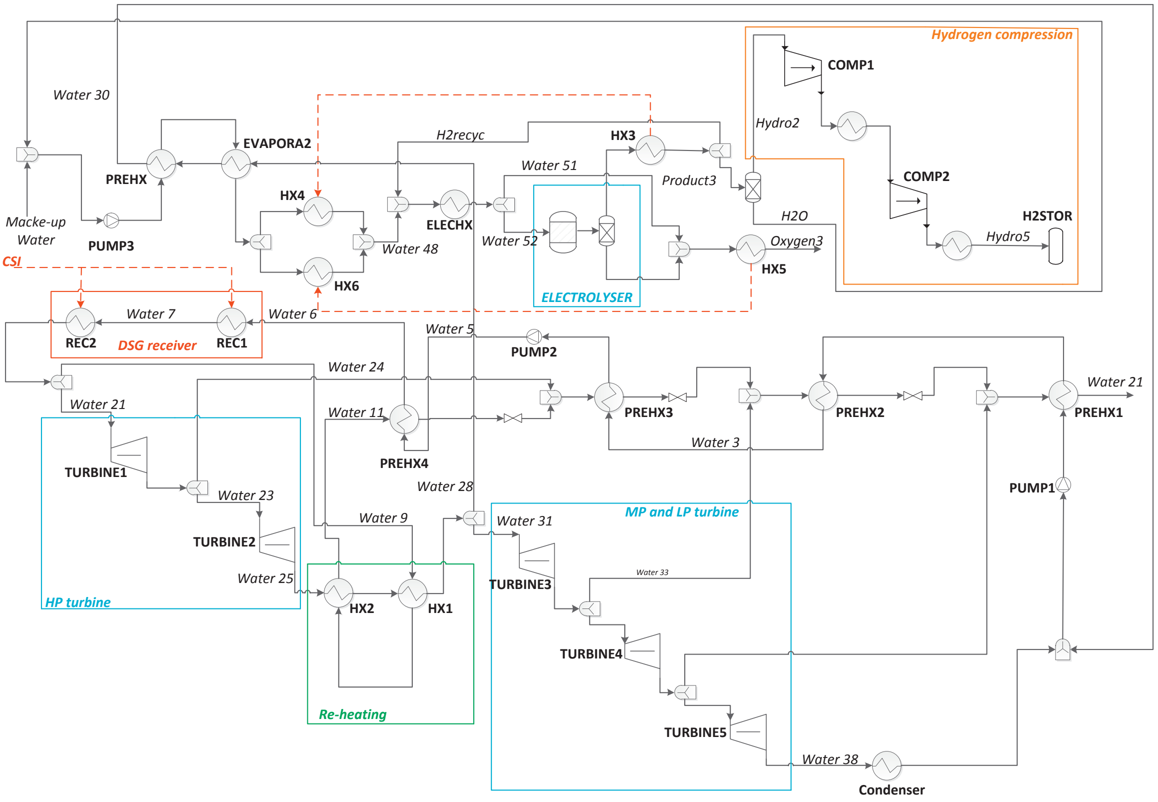

Fig. 5. Flowsheet of overall process for solar high-temperature electrolysis.

The high concentrated solar radiation is used in the solar receiver to generate steam at the fixed conditions of 550°C and 160 bar. The solar receiver was simulated as two high temperature heat exchangers; the first heat exchanger REC1 aims to the evaporation of the preheated water WATER6 and the superheating of the steam WATER7 is performed in the second heat exchanger REC2. A part of the superheated steam WATER9 is extracted from the solar receiver and further used for the reheating process. The other part of the steam WATER21 is introduced to the first high pressure steam turbine TURBINE1. After expansion, a part of the steam WATER24 is splitted and used as heat transfer fluid in the preheater PREHX3in order to preheat the water stream WATER3. The other part of the steam WATER23 is expanding in the second high pressure steam turbine TURBINE2. After the second expansion, the steam WATER25 is reheated up to 335°C in the heat exchangers HX1 and HX2 by the stream WATER9 extracted from the solar receiver. After reheating, the steam WATER11 is used to preheat the feed water WATER5 in the heat exchanger PREHX4. The heat transfer fluid for the evaporation of the HTSE feed water is steam generated in the solar receiver, which is splitted after the reheating process. The other part of the steam WATER31 is expanding in the medium steam turbine TURBINE3. Further expansion of the steam is performed in the low pressure turbines TURBINE4 and TURBINE 5. After leaving the last turbine, the steam WATER38 is condensed and reintroduced to the process with the water stream WATER30a, which has been used for the evaporation of the HTSE feed water.

Exhaust lines of the HTSE stack are coupled to the heat recovery system, which consists of two high temperature heat exchangers HX3/HX4 and HX5/HX6 where the HTSE feed steam is heated up to 634°C. Each high temperature heat exchanger was simulated as two single heat exchangers, which are combined with a heat stream. An electrical heater ELECHX2 is required in order to superheat the steam up to the operating temperature of the electrolyzer of 700°C. In the HTSE stack, steam is supplied to the cathode side and is splitted into hydrogen and oxygen ions. The oxygen ions are transferred through the electrolyte to the anode side where they combine to form oxygen. The overall reaction is given by equation 1. The steam supplied to the cathode side is first mixed with a part of the produced hydrogen in order to have a composition of 90%mol H_2O and 10%mol H_2. A steam to hydrogen conversion of 60% was assumed based on the results of the EU founded project ADEL. The product gas containing hydrogen and water PRODUCT3 is introduced to the flash unit, where the steam is separated by condensation and then introduced again as HTSE feed water. The hydrogen stream HYDRO 2 is compressed up to 30 bar in a two stages compressor. The next table summarizes the power generated by the steam turbines and the energy consumptions of core components.

Table 2. Simulation results of core components

Components	Power [MW]
Turbines	
TURBINE1	17.56
TURBINE2	7.75
TURBINE3	11.03
TURBINE4	7.05
TURIBNE 5	8.34
Solar receivers	
REC1	113.22
REC2	66.4

The results of the main streams of the process have been summarized in the next section.

Table 3. Results of the main streams of the process

Stream	Mass flow [kg/h]	Mole flow [kmol/h]
Rankine cycle feed water	$2.77\ 10^5$	15386.94
HTSE feed water	4859.58	270.01
Sweeping steam	4320.16	270.01
Hydrogen production rate	409.63	168.85
Oxygen production rate	2688	85.37

The efficiency of the Rankine cycle can be defined as:

$$\eta_{th} = \frac{\dot{W}_{turbine} - \dot{W}_{Pumps}}{\dot{Q}_{in}} \tag{9}$$

η_{th} defines the thermodynamic efficiency of the cycle as the ratio of net power output to heat input. The thermal energy used for the generation of superheated steam is about 154.7 MWth since a part of the total thermal energy absorbed by the solar receiver is introduced to the HTSE process to evaporate the feed water of the electrolyzer.. The total electrical power generated by the HP and LP turbines is about 51.73 MWel. The work required by the pumps is around 1% of the turbine work output and the equation 9 can be simplified. As a result, the thermodynamic efficiency of the Rankine cycle is about 33.43%.

The overall efficiency of the HTSE process coupled to the DSG power plant can be defined as the ratio of the energy carried by unit amount of produced hydrogen, in terms of high-heat value of hydrogen (HHV=285.8kJ/mol), to the overall energy input of the process, which is in this case the electricity input of the electrolyzer. Indeed, the HTSE process is characterized by energy losses related to the ohmic loss due to resistance of SOEC medium to electron and ion transfer. The considered HTSE process in this paper is ideal. Therefore, the ohmic losses will not be taken into consideration for the calculation of the efficiency.

The electrolyzer efficiency of the plant can be expressed as:

$$\eta_{overall} = \frac{\dot{n}_{H_2}\, HHV_{H_2}}{P_{el}} \tag{10}$$

According to the simulation results, the electricity input of the electrolyzer corresponds to the power generated by the steam turbines of 51.73 MWe producing a hydrogen flow rate of 168.85 kmol/h. As a result, the efficiency of the HTSE process is about 26 %**.** The overall efficiency of the process can be increased by increasing the steam-to-hydrogen conversion, which has been set at 60 % in this study. Furthermore, it was assumed that 500 kWe are required for the electrolysis of 1.63 kmol/h of water. This assumption regarding the capacity of the electrolyzer could be modified. In order to have an idea about the impact of the steam to hydrogen conversion and the capacity of the electrolyzer, a sensitivity analysis has been carried out. Regarding the long term development in the field of high temperature electrolysis cells, the steam-to-hydrogen conversion and the capacity of the electrolyzer are key factors for the improvement of the efficiency. Following assumption has been made to study the impact of increase in conversion on the efficiency: by assuming a steam-to-hydrogen conversion of 90% instead of 60%, the efficiency of the HTSE process increases from 26% to 38%. By assuming an increase in the capacity of a 500kWe electrolyzer from 1.63 kmol/h up to 3.26 kmol/h, the efficiency increases from 38% to 47% by maintaining the steam to hydrogen conversion at 90%.

5. Conclusions

A tube-type receiver has been developed and tested in the solar simulator of DLR in Cologne to superheat steam from 100 °C to 700 °C for use in a high temperature electrolyzer process. The receiver is constructed of 20 absorber pipes made of high alloy steel assembled in a concentric, cylindrical configuration forming a circular aperture with a diameter of 90 mm. In a layout study based on heat balancing and ray tracing the tube length was set to 270 mm. After a successful initial operation of the system a series of test runs were conducted superheating steam with a flow rate of up to 5 kg/h to an outlet temperature of 700 °C achieving a thermal efficiency of 40 % with about 4 kW of solar power. Higher mass flows will be tested in upcoming experimental campaigns. In a possible follow-up project the receiver is planned to be coupled to an electrolyzer stack to demonstrate the integrated operation of a solar HTE. In addition to this experimental work, a flowsheet of a process for solar high temperature electrolysis driven by a central receiver system for direct steam generation was designed. In a steady-state simulation an efficiency of 33 %

was found for the Rankine cycle included in the process. The conversion of thermal energy to hydrogen occurred at an overall efficiency of 26 % considering a rather low steam to hydrogen conversion of 60 %. However, high temperature electrolyzers have the potential to achieve significantly higher steam-to-hydrogen efficiencies of about 90 %. Taking this into consideration in a sensitivity analysis the thermal-to-hydrogen efficiency increased to 38 %. Even higher values of up to 47 % could be reached at a higher electrolyzer capacity. In a next step an optimization of the steady-state operation will be carried out followed by a transient simulation of the process including ray tracing of the solar field. Finally, the flowsheeting analysis will form the basis of a techno-economic study of a high temperature electrolysis process with direct steam generation.

Acknowledgements

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