

Economic comparison of solar hydrogen generation by means of thermochemical cycles and electrolysis

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ABSTRACT

Hydrogen is acclaimed to be an energy carrier of the future. Currently, it is mainly produced by fossil fuels, which release climate-changing emissions. Thermochemical cycles, represented here by the hybrid-sulfur cycle and a metal oxide based cycle, along with electrolysis of water are the most promising processes for 'clean' hydrogen mass production for the future. For this comparison study, both thermochemical cycles are operated by concentrated solar thermal power for multistage water splitting. The electricity required for the electrolysis is produced by a parabolic trough power plant. For each process investment, operating and hydrogen production costs were calculated on a 50 MW_{th} scale. The goal is to point out the potential of sustainable hydrogen production using solar energy and thermochemical cycles compared to commercial electrolysis. A sensitivity analysis was carried out for three different cost scenarios. As a result, hydrogen production costs ranging from 3.9–5.6 €/kg for the hybrid-sulfur cycle, 3.5–12.8 €/kg for the metal oxide based cycle and 2.1–6.8 €/kg for electrolysis were obtained.

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1. Introduction

As one of the most promising future energy carriers, hydrogen is currently produced from fossil resources using reforming or gasification processes. This leads to carbon dioxide (CO₂) emissions of $0.3-0.4 \text{ m}^3\text{CO}_2/\text{m}^3\text{H}_2$ [1] and thus augments the greenhouse effect. In fact, hydrogen can only be considered as an environmentally friendly and sustainable alternative to fossil energy carriers, if it is produced from renewable energy and without harmful emissions. Thermochemical cycles (TCC) and electrolysis of water are environmentally friendly and most promising alternatives for the long-term CO₂-free hydrogen production, if operated by concentrated solar power. Both will be explained in detail and compared concerning their economic efficiency (investment, H₂ output, H₂ production costs) subsequently. The calculations of heat balances, solar field size and shape were made for plants with an annual average thermal power of 50 MW at a suitable site. The measure for the scale of the plant is the amount of heat coupled into the process. Costs for hydrogen compression, storage and distribution are not considered in this study.

2. Process description and plant layout

TCCs are processes which decompose water into hydrogen and oxygen via chemical reactions using intermediate reactions and substances. All of these intermediate substances are recycled within the process. Thus, the sum of all the reactions is equivalent to the dissociation of the water molecule. Theoretically, only heat is necessary to process these chemical steps [2]. This can be provided by concentrated solar energy using a central receiver system (CRS). The CRS consists of mirrors, so-called heliostats, which concentrate the sunlight

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NomenclatureAFannuity factor CO_2 carbon dioxideCRScentral receiver systemCSPconcentration solar powerDLRGerman Aerospace CenterDNIdirect normal irradiation e^- electronH_2SO_4sulfuric acidHPChydrogen production costs i_r interest rateLEClevelized electricity costs	$\begin{array}{llllllllllllllllllllllllllllllllllll$
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onto the receiver. The receiver located on the top of the tower is made up of honeycomb-structured ceramics acting as an absorber for the solar radiation and as a chemical reactor.

Solar thermochemical hydrogen generation based on metal oxides and the hybrid-sulfur cycle are among those few candidates having the highest potential in terms of the feasibility of scale-up [3]. On the contrary, electrolysis, which is the electrochemical decomposition of water into hydrogen and oxygen [4], is an already mature technology which is seen as a benchmark for future hydrogen production.

Altogether, thermochemical cycles have the potential of a higher efficiency than alkaline electrolysis [3] and hence have the potential to reduce the production costs of hydrogen from water significantly.

2.1. Thermochemical hydrogen production based on metal oxides

The first considered process is a two-step thermochemical cycle based on metal oxides (MO) serving as the catalyst. The reaction scheme is as follows:

 $MO_{reduced} + H_2O \rightarrow MO_{oxidized} + H_2(splitting 800 \,^{\circ}C)$

 $MO_{oxidized} \rightarrow MO_{reduced} + O_2(regeneration 1200 \,^{\circ}C)$

The metal oxide acts as redox system which is fixed on the surface of a porous absorber. One promising redox system is the class of mixed iron oxides, as investigated, e.g. within the EU-Project HYDROSOL I + II, [5,6]. At the beginning, the metal oxide is present in a reduced form. By adding water vapor at 800 °C, oxygen is abstracted from the water molecules and hydrogen is produced. When the metal oxide system is saturated - meaning fully oxidized - it is heated for regeneration at 1100 °C to 1200 °C in an oxygen-lean atmosphere. Oxygen is exhausted from the redox system using nitrogen as a flushing gas [5]. The product gas passes through heat exchangers and is cooled down before residual water is separated. The receiver surface is divided into several square apertures; two apertures make up one receiver pair (Fig. 1). One aperture is applied for the dissociation of the water vapor, while the other one is used for the regeneration of the redox system. Thus, hydrogen can be produced continuously by alternating the reaction steps.

2.2. Hybrid-sulfur cycle

The hybrid-sulfur cycle, occasionally called "Westinghouse Cycle" [7], was investigated within the EU-project HYTHEC [8]. It is based on the decomposition of sulfuric acid (H_2SO_4) as a high temperature thermochemical step and the low temperature electrolytic reaction of water and sulfur dioxide (SO_2) [9]. The high temperature step consists of two sections: initially H_2SO_4 is thermally decomposed into water vapor and sulfur trioxide (SO_3) and the latter is reduced at temperatures up to 1200 °C to SO_2 and oxygen. Oxygen is separated and might be used as a byproduct.

In the low temperature step, SO_2 is oxidized to H_2SO_4 at the anode of an electrolyzer, while hydrogen is formed at the cathode according to the reaction scheme [10]:

 $\mathrm{H_2SO_4} \rightarrow \mathrm{H_2O} \,+\, \mathrm{SO_3} \rightarrow \mathrm{SO_2} \,+\, {}^{1\!\!/}_{2}\mathrm{O_2} \,+\, \mathrm{H_2O}$

(thermal decomposition 1200 °C)

 $2H_2O\ +\ SO_2 \rightarrow H_2SO_4\ +\ H_2(electrolysis\ 85\ ^\circ C)$

For this process, a solar tower could consist of two cylindrical apertures; one for the electrical power generation and the other one for the chemical receiver reactor (Fig. 4). A helium-based Brayton cycle¹ could also be used for power generation. After the decomposition of the sulfuric acid, the hot gases flow through heat exchangers to transfer the excess heat to incoming streams. Oxygen is separated from the product gas flow by absorber units. Water is added to the cycle in the electrolyzer to oxidize the purified SO₂ to H_2SO_4 and hydrogen.

2.3. Electrolysis

Water electrolysis means the electrochemical splitting of water into oxygen and hydrogen using a conductive electrolyte (salts, acids, bases). In this case, an alkaline electrolyte is considered. Hydrogen is generated at the cathode while oxygen is produced at the anode of the electrolyzer. The charge equalization proceeds by ionic conduction. A porous membrane is located between the two electrodes to prevent the mixture of the product gases. To be a CO_2 -free

¹ The Brayton cycle is a cyclic process generally associated with the gas turbine.



Fig. 1 – Aperture design of receiver clusters (metal oxide based cycle), cross-sectional view.

production process, electricity required for electrolysis must be generated by renewable energies. For this evaluation, electricity production by concentrated solar energy using a parabolic trough power plant was assumed. Electrolysis of water has been commercially used for over 80 years and is particularly suitable for locations with low-cost electricity [11].

 $2H_2O + 2e^- \rightarrow H_2 + 2OH^-(cathode)$

 $2OH^- \rightarrow \frac{1}{2}O_2 + H_2O + 2e^-(anode)$

3. Design parameters and method of calculation

The average annual power of the CRS and parabolic trough power plant is 50 MW_{th} in each case. The performance of solar energy plants strongly depends on their sites and the corresponding insolation conditions. Region suitable for massive hydrogen production by concentrated solar energy is preferably within the sunbelt along the tropics. Besides a high average direct normal irradiation (DNI), a good location offers sufficient access to fresh water as well as an adequate infrastructure for hydrogen transportation. The region of Assuan close to Lake Nasser (Egypt) fulfills all criteria and was, therefore, chosen for this examination. The annual irradiation is 3137 kWh/(m² y) during 4214 h of sunshine [12]. The design point of the plant is June 21st at noon.

For the heliostat field layout, the simulation tool HFLCAL was used [13]. Based on the chosen configuration, the annual hydrogen and electricity output was calculated. Then by means of the resulting mass flows the plant components were designed. The main focus within the economical evaluation is the cost comparison of Total Capital Investment (TCI), cost of Operation and Maintenance (O&M) as well as Hydrogen Production Costs (HPC).

The TCI and O&M costs are calculated based on results of simulation, literature and requested quotes. Also, a safety surcharge of 10% [14] is added to the Total Direct Investments (TDI) of components to cover the costs of engineering, construction and contingencies. The TCI results from the addition of TDI (components) and Total Indirect Investment (infrastructure) of the plant.

In all cases a lifetime (n) of 20 years and interest rate (i_r) of 6% are assumed. The O&M costs reflect fixed and variable costs per year, added for the whole period of consideration. Besides hydrogen, in all considered processes the by-product oxygen is produced and its revenue (R_{O_2}) offsets the O&M costs.

Finally, the HPC is calculated using the annuity method. At first the present value (PV) was defined using the calculated costs and revenue. Multiplication of PV with the Annuity Factor (AF) establishes an annual basis, so that finally the division with the annual amount of produced hydrogen ($P_{\rm H}$) gives the HPC (Eqs. (1)–(3)).

$$PV = TCI + \sum_{n=1}^{n_p} \frac{(O\&M)_n - R_{O_{2n}}}{(1+i_r)^n}$$
(1)

$$AF = (1 + i_r)^n \frac{i_r}{(1 + i_r)^n - 1}$$
⁽²⁾

$$HPC = \frac{PV \times AF}{P_{H}}$$
(3)

4. Commercial plant design and cost estimates

Cost estimation is based on results of simulations and experiments within the mentioned projects as well as by published information. The following general assumptions have been made for economic analysis:

- A CRS was chosen for heat supply of both TCCs.
- Heliostat costs are assumed with $120 \in /m^2$ (164 $/m^2$) [15].
- Electricity required for the metal oxide based cycle and electrolysis is provided by a parabolic trough power plant.
- The required feedstock and other consumables are available on site.
- Costs for the land are assumed for all cases with 2 M€/km² [16].
- Oxygen as a by-product is sold for 0.15 €/m³.

O&M costs are composed of labor, consumables and repairs/replacements. Supervision and operation is done by 15 employees, each assumed to cost 48 k€ per year being representative of the Mediterranean region as described in the ECOSTAR study [16]. The costs for insurance and maintenance are defined to be 2% and 4% of the TCI, respectively [17]. Thus, increasing costs for maintenance over the entire term remain unconsidered. Demineralized water as feedstock of hydrogen is fed into the process for $1.09 €/m^3$ in each case [18]. Electricity is obtained for 0.1 €/kWh for the electrolysis and the metal oxide-based process [16].

Further assumptions and determinations concerning the installation and costs of the equipment will be explained subsequently for each process and summarized in Table 1.

4.1. Economic parameters of the metal oxide cycle

For solar thermochemical hydrogen generation based on metal oxides, a field of 3199 heliostats each with a reflective surface of 121.34 m² is required. They are arranged around a 176-m-high tower made of reinforced concrete (costs 2.8 M€). The receiver on the top of the tower has 16 apertures, which are uniformly distributed around the perimeter of the tower top. Between the individual receiver surfaces, preheating zones for the incoming gas flows are arranged (Fig. 1). The entire receiver surface amounts to 384 m² and costs 4.2 M€. Due to the fragmentation of the receiver area the heliostat field is subdivided into eight different sub-fields in which the respective mirrors aim at their dedicated aperture (Fig. 2). The sizes of the sectors depend on the position of the sun and position of the heliostats, so that every sector yields the same annual power. Heliostats, tower and receiver, make up the Solar Equipment. Afterwards the heat of the product gases is transferred to the input flows within heat exchangers preferably located on the top of the tower also. Then the flow runs to the bottom of the tower where it is continuously cooled down for subsequent separation steps. Chemical application includes costs for cooling (5 M \in), compression (1.5 M \in) and heat exchangers (10 M€).

The nitrogen required for the regeneration of the redox system is fed into the process from a separate storage tank. The capacity of the tank is designed to allow three complete H_2 -production cycles per run (volume 180,000 m³; costs 1.3 M \in). On the commissioning date, the tank has to be filled with nitrogen and was, therefore, assigned to the TCI (costs 0.182 \in /m³) [19]. Costs for pumps, piping and connection of components are assumed to be 28% of the overall chemical



Fig. 2 – Design of the heliostat field (metal oxide based cycle).

plant investment (chemical application and tank), based on estimations of Timmerhaus [14]. For solar equipment, this charge is already included in the investment. The land covered by heliostats is 1.7 km^2 . The area directly around the tower, where no heliostats can be placed for optical reasons, can be used for the chemical plant components. To protect the components against the impacts of atmospheric conditions, they will be placed in a building, assumed to be 1900 m² at $600 \notin m^2$ [20].

In addition to the costs of employees, insurance and maintenance, 37,350 $\rm m^3$ of water and 97.5 GWh_e are needed for operation. Due to the fact that the redox system has to be

Table 1 – Technical and economic data							
	Unit	Thermochemical cycle based on metal oxides	Hybrid-sulfur cycle	Electrolysis of water			
Technical data							
State-of-the-art		Pilot plant	Lab scale experiments	Industrial modular			
Power		100 kW _{th}					
Technological maturity 2025–2030		Prototype, commercial	Demonstration, commercial	Matured, commercial			
CO ₂ level		Free	Free	Depending on the electricity generation method			
H ₂ production	[t/y]	4150	3740	2464			
Receiver size	[m ²]	384	132 (TCC) 88 (electricity generation)	-			
Economic data							
Heliostats (121.34 m²)	[€/m²]	120	120	-			
Land	[M€/km²]	2	2	Contained in LEC			
Safety surcharge	[% of TDI]	10	10	10			
Water input	[m³/y]	37,350	33,660	31,307			
Electricity input	[Gwh _{el} /a]	97.5	-	139.9			
Redox system	[t/y]	16.3	-	-			
Nitrogen (input & recycling)	[M€/y]	5.5	-	-			

substituted at a loss of more than 20%, the corresponding costs will be assigned to O&M too. An amount of 16.3 tons of ferrites is required annually. As the redox system is custommade, the potential of cost reduction is huge. From currently 1500 €/kg [21] produced in small laboratory scale, prices of 150 €/kg are projected after market launch. Finally, it is assumed that the price will decrease to 15 €/kg in the remote future. Such price margin and price development is known from costs of other chemicals like titanium dioxide (TiO₂). TiO2 is used in several applications, e.g. as component of toothpaste, wall paint or as a catalyst. While it is sold for 1230 €/kg [22], when purchasing few grams for laboratories, it is sold for only 15 €/kg to industrial consumers [23]. Due to the fact that the price of the raw materials and the production process is in some respect similar to the ferrite redox system, it is assumed that the price difference will be almost equivalent. Thus, costs of 150 €/kg ferrite are taken as the standard case, while the other extreme values will be considered in the sensitivity analysis subsequently.

Small losses of the flushing gas during the regeneration step were calculated to be 0.1% of the collecting main capacity per year, which equals 32,184 m³. They are compensated by supply from the tank system. Furthermore, the separation of nitrogen and oxygen is estimated to cost $0.017 \in$ per recycled m³ N₂ [19]. The annual output of the plant is calculated to be 4150 tons H₂ during 4053 h of operation. The TCI is 89.0 M \in and the costs for O&M amounts to 23.8 M \in per year (Table 2).

4.2. Economic parameters of the hybrid-sulfur cycle

The plant for solar thermochemical hydrogen generation by hybrid-sulfur cycle consists of 2627 heliostats (Fig. 3). The needed electric and thermal energy are both provided by separate receivers on a 200-m-high solar tower (Fig. 4). Investment for heliostats (38.2 M \in), tower (3.7 M \in) and receiver (5.2 M€) add up to be 47.1 M€. The receiver for electricity generation is designed as a tube receiver with a receiver area of 88 m^2 . The H₂SO₄ decomposition takes place within several volumetric receivers consisting of honeycombstructured ceramics with a surface of 132 m². Downstream of the volumetric receiver the heat of the gas flow is transferred by heat exchangers. The gas stream leaving the decomposition unit is separated into pure O₂ and SO₂ which is dissolved in water by absorption columns. Investment for chemical application includes heat exchangers (16.8 M \in), cooling (8.2 M \in), separation (1.7 M \in), pumps and compression (2.2 M \in).

Due to the lack of SO₂-electrolyzers, an alkaline electrolyzer acts as reference for design and costs [24]. Characteristics and costs were calculated within the HYTHEC project [9]. Electrodes are assumed to be made of carbon steel with a nickel coating the membranes of an asbestos-free polymer material. The reference electrolyzer considered here includes 230 cells each with a production rate of 43.6 kgH₂/h. In addition an overhead cost for the platinum deposit has been considered in this analysis. All in all, 14 electrolyzers are used adding up to 38.5 M \in .

Table 2 – Composition of investments (M \in) and operating costs (M \in /a)						
		Thermochemical cycle based on metal oxides	Thermochemical cycle hybrid-sulfur	Electrolysis of water		
Direct investment	Solar equipment	53.6	47.1	-		
	Chemical application	16.5	29.1	-		
	Tanks	1.5	-	-		
	Electrolyzer plant	-	38.5	6.0		
	Electricity generation	-	6.3	-		
	Pumps, piping, connection	5.0	13.3	1.7		
	Buildings	1.1	1.8	0.4		
Indirect investment	Safety surcharge	7.8	13.6	0.8		
	Sulpuric-acid	-	0.02	-		
	Nitrogen	0.04	-	-		
	Land	3.4	2.2	-		
Total capital investment (Mel	89.0	152.0	8 9		
Fixed costs	Mannower	0.7	0.7	0.7		
	Insurance	1.8	3.0	0.2		
	Maintenance	3 5	6.1	0.2		
	Redox-system	2.5	-	-		
Variable costs	Electricity	9.8	_	14 3		
	Water	0.04	0.04	0.03		
	Nitrogen (feed)	0.006	-	-		
	Nitrogen (recycling)	5.5	-	-		
		00.0	0.0	45.6		
Total O&M costs [M€/a]		23.8	9.9	15.6		
By-product credits [M€/a]		3.5	3.1	2.1		
H_2 production costs [\in /kg]		6.7	5.4	5.8		
Specific TCI [M€/MWth]		1.8	3.0	0.2ª		
Specific O&M [M€/a MW _{tl}	h]	0.13	0.20	0.31		
a Investment of the parabolic trough power plant is included in electricity costs.						



Fig. 3 - Design of the heliostat field (hybrid-sulfur cycle).

The required electricity shall be completely generated by a helium-based closed Brayton cycle using solar thermalgenerated steam. This concept was chosen because of its high efficiency and good part-load characteristics. Economic data for the closed Brayton cycle are not published in detail yet. To get a rough estimate, the costs for open gas turbine systems ($6.3 M \in$) are assumed [25]. This is justified by the fact that on one hand, the casings of the closed systems are heavier and thus more expensive due to the higher pressure level, on the other hand, very expensive features required for open gas turbines (combustion chamber, coating and cooling) can be



Fig. 4 – Tower with two annular receiver apertures, one for power generation and one for H₂SO₄ decomposition (hybrid-sulfur cycle), cross-sectional view.

omitted due to extremely high temperatures. In total a power level of 12.51 MW_{e} is required.

Based on the separated-receiver concept, the plant can be operated optimally depending on the thermal power that is available at a certain time. For this purpose the heliostats are switched between the receivers. Therefore, no tanks are required for the chemicals. Sulfuric acid circulates within the system and sulfur dioxide is directly disposed to the electrolyzer. Due to the fact that sulfuric acid has to be in the plant for start-up, the costs are also regarded as an (indirect) investment. It is assumed that 50 tons of H₂SO₄ are needed where the price is given with 300 \in /t.

Within the open space between tower and heliostats all components are placed on 3050 m^2 and are roofed by a light metal construction calculated with $600 \notin m^2$ [20]. The investment for piping and connection are defined with 18% of the component investment (chemical application, electrolyzer and electricity generation) [14]. Pumps were already considered separately in the chemical applications. The whole power plant covers a surface area of about 1.1 km².

Aside from fixed operation costs, 33,660 m³ of water must be supplied annually. Additional chemicals are not required and electricity is completely generated by the closed gas turbine process. The annual hydrogen production is 3740 tons during 4089 h of operation. The TCI of the plant is 152.0 M \in and the annual O&M costs amount to 9.9 M \in (Table 2).

4.3. Economic parameters of the electrolysis

The evaluation of electrolysis is based on costs of solar generated electricity by parabolic trough collectors and the investment for the alkaline electrolysis cells. More precisely, no investment and O&M will be calculated for the solar field, but the cost and the amount of electricity generated by a solar power plant with a storage time of 4.5 h and an annual average power of 50 MW_{th} will be available for electrolysis. The annual net electricity output of the solar power plant is calculated to be 139,846 MWh considered with 0.1023 \in /kWh_{el} [16]. The Norwegian company Hydro offers electrolyzers requiring 4.3 kWh_e/m³ H₂ (47.8 kWh_e/kg) which is equivalent to an efficiency of 70% [24]. Overall four of these electrolyzers each with 1.5 M [26] and a heat storage capacity of 420 MWh_{th} are required. The investment of the thermal storage is already included in the electricity costs [16].

Costs for pumping, piping and connection are covered by 28% of the electrolyzer investment. The comparable small land area required is included in the cost of buildings (0.4 M). The land required for the CSP installation is already contained in the LEC [16]. Operating costs for manpower, insurance and maintenance are calculated as described above. Additionally 31,307 m³ of demineralized water must be supplied annually to produce a total of 2464 tons of hydrogen. All in all, the TCI of the electrolyzer plant is 8.9 M€ and O&M amounts to 15.6 M€ per year (Table 2).

5. Results

The results of the economic analysis are presented in Tables 1 and 2. Electrolysis clearly shows the lowest investment costs and the lowest hydrogen output at the same time. This is because only the

electrolyzer is counted among the TCI whereas operation costs are mostly influenced by electricity costs, which are considered instead of the investment of a parabolic trough power plant. The resulting HPC of a 50 MW_{th} plant is calculated to be 5.8 \in /kg.

Compared to that, investment of the hybrid-sulfur cycle is almost 17-times higher due to the required complex chemical plant, mostly made out of corrosion-resistant materials, and the thermo-electric system. Due to the integration of the Brayton cycle, all required electricity is generated by the system itself. Thus O&M costs of the hybrid-sulfur cycle are the lowest. Due to the relation of relatively high investment and low O&M costs, the HPC are $5.4 \notin$ /kg.

The metal oxide based cycle shows the highest O&M costs. This is because the redox system must be renewed periodically and the price is based on a custom-made product. Also, the flushing gas nitrogen has to be replaced and separated constantly. However, the metal oxide based cycle produces the biggest amount of hydrogen with an HPC of $6.7 \in /kg$.

The HPCs obtained for the considered processes seem to be unfavorable compared to fossil energy based processes. However, the advantage, apart from the ecological benefits, is the permanent presence and infiniteness of solar energy. Also, the potential of cost reduction for components and materials after market launch is quite high as shown in the following sensitivity analysis.

6. Sensitivity

The previous cost estimation has been made using currently available data and mainly based on custom-made components and materials. These costs will definitely decrease within the next years due to technological progresses and commercialization. This sensitivity analysis presents three different cost scenarios: conservative, standard and optimistic (Table 3). The conservative scenario mainly includes the current costs. On the other hand, the optimistic case refers to the prices, which seem achievable for commercial facilities. For the standard case, prices are assumed which can be realized by market launch and mass production.

For heliostats, costs are currently influenced by increasing costs of steel and the demand. Nevertheless, an optimistic $100 \notin m^2$ could be attained in the future using technology

Table 3 – Overview of parameters for sensitivity					
Criteria	Cost scenario				
	Conservative	Standard	Optimistic		
Heliostat costs [€/m²]	140	120	100		
Lifetime [a]	20	25	30		
Redox system costs [€/kg]	1500	150	15		
Electricity costs [€/kWh _{el}]	0.12	1.10	0.04		
Electrolyzer [decrease in %]	0	-10	-20		
Chemical application [decrease in %]	0	-10	-20		
Recycling of nitrogen [decrease in %]	0	-20	-40		

improvement opportunities [15]. As mentioned in Section 4.1, the reduction potential of the metal oxide is immense. The price ranges from 15 (commercial, mass production) to $1500 \in$ /kg (custom-made). Electricity required for the metal oxide based cycle and electrolysis have the potential to decline to $0.04 \in$ /kWh until the year 2020 [16]. Even though it seems unlikely that costs of solar generated electricity will increase in the future, they are considered to be $0.12 \in$ /kWh as the conservative scenario. The operating time of 20 years could be extended to 25 years (standard) or 30 years (optimistic) by technological progress and improvement of durability.

The existing costs for electrolysis are assumed to be reduced by 10% (standard) to 20% (optimistic) in the future [26]. That applies for the chemical application too. Also, the N₂ recycling has an influence on the HPC of the metal oxide based cycle. Further investigations and process simulations are necessary for optimization. The goal is to replace the flushing gas nitrogen by a vacuum pump for the elimination of oxygen. This in turn implicates a modification of the receiver design and heliostat field as well as additional costs. First estimations indicate the creation of a more favorable situation by these means.

Finally, the results of the sensitivity study are shown in Fig. 5. All optimistic, standard and conservative costs of each parameter are summarized in the column 'Best, Standard and Worst Case'.

As a result, variation of the chosen parameters has the least impact on the HPC of the hybrid-sulfur process (variation from 3.9 to 5.6 ϵ /kg). The main cost factor of electrolysis is the electricity: just the variation of electricity costs leads to HPC of 2.1–6.8 ϵ /kg. Additionally reduction of electrolyzer costs and the extension of operating time lead to a minimum HPC of 2.1 ϵ /kg. The maximum range of HPC is obtained at the metal oxide based process with 3.5–12.8 ϵ /kg. As discussed before, the redox system has the largest impact on the HPC for the metal oxide based cycle. The variation yields HPCs of 6.2–12.1 ϵ /kg. The high electrical energy demand for nitrogen recycling influences the result significantly.

All of the current mass production processes depend on fossil fuels and thus lead to harmful emissions. In the future, a rise in prices due to the shortage of resources as well as CO₂ fines is likely. Currently, most of the hydrogen is produced by natural gas steam reforming for about 6.5 \in /GJ (equivalent to 0.8 \in /kg) [27] and coal gasification with 0.065 \$/kWh_{th} (1.8 €/kg) [28]. Projected primary energy costs in 2020 are 7.06-9.41 €/GJ (0.85-1.13 €/kg) for natural gas reforming and 8.44–11.70 €/GJ (1.01–1.4 €/kg) for coal gasification without carbon capture costs [27]. Provision and implementation of carbon capture technology (without transport and storage) will lead to an increase in HPC by 20-25% [27]. Indeed, HPC of the TCCs are higher than the fossil costs at this time; however, TCC have the potential to achieve lower production costs with new technology whilst the reforming and gasification processes strongly depend on the development of fuel prices. Then CO₂-free hydrogen production processes will be more competitive and enable the transition to hydrogen as an environmentally friendly energy carrier.

7. Conclusion and outlook

Based on flow sheets and simulations, investment and operating costs have been determined for the economic



Fig. 5 – Sensitivity of hydrogen production costs by three production processes and a conservative, standard and optimistic price scenario.

comparison of three CO₂-free hydrogen production processes. Representative for this examination are two thermochemical cycles, one based on metal oxides and the hybrid-sulfur cycle, compared with electrolysis of water using solar generated electricity. The values provide a basis for the calculation of the hydrogen production costs using the annuity method. The potential for cost reduction is huge, which was shown in the sensitivity analysis for three cost scenarios. The hybridsulfur cycle shows the lowest HPC for the calculated standard case (5.4 €/kg) and has the smallest cost range (3.9–5.6 €/kg). For water electrolysis, the HPC varies between 2.1 and 6.8 €/kg which is mainly influenced by the high demand and costs of electricity. For the future, stability and lifetime of the electrolyzer should and will be the most important targets for improvement. Nevertheless, electrolysis can only be competitive in regions with low electricity costs. The metal oxide based cycle will yield to HPC in a range between 3.5 and 12.8 €/kg, mainly caused by the high demand of the metal oxide, which is custom-made today but will be mass produced if applied in large scale. In contrast to the hydrogen production processes, which are currently applied on a commercial scale, the three investigated processes do not produce any harmful emissions. Indeed, steam reforming of natural gas is currently the most favorable hydrogen production method from a techno-economic point of view, but a major concern is the increase of natural gas price [29].

The first plants for CO₂-free hydrogen mass production are projected to start between 2020 and 2025. Under consideration of commercialization and batched flow production of components, 'clean' hydrogen production can be competitive soon.

Thermochemical cycles attract attention around the world. Hydrogen production using ferrites is now in the demonstration state. Within the EU HYDROSOL II project, a $100 \text{ kW}_{\text{th}}$ pilot plant will be installed at Plataforma Solar de Almería by 2008 [5,6,30]. This test campaign aims at an optimization of efficiencies as well as lowering the temperature level of regeneration step considerably below 1200 $^\circ\text{C}.$

The European project HYTHEC was completed by the end of 2007. Several test campaigns have been carried out at the DLR solar furnace in Cologne, where the general operability of the reactor concept and a homogenous H_2SO_4 decomposition has been proven. For the future, it is essential to enhance, scale-up and demonstrate the sulfuric acid step at pilot plant scale. Furthermore, the development of efficient electrolyzers has to be promoted. In addition, the development, qualification and certification of resistant materials for the components and the coupling of solar or nuclear heat will be a main task in the future. This work will be done within the project HycycleS which has already begun in January 2008 [31].

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REFERENCES

- Muradov N. Hydrogen via methane decomposition: an application for decarbonization of fossil fuels. Int J Hydrogen Energy 2001;26:1165–75.
- [2] Funk JE. Thermochemical hydrogen production: past and present. Int J Hydrogen Energy 2001;26:185–90.

- [3] Kolb GJ, Diver RB. Screening analysis of solar thermochemical hydrogen concepts. Sandia Report Sand2008-1900. Unlimited Release; Printed March 2008.
- [4] Barbir F. PEM electrolysis for production of hydrogen from renewable energy sources. Sol Energy 2005;78:661–9.
- [5] Roeb M, Sattler C, Klüser R, Monnerie N, de Oliveira L, Konstandopoulos AG, et al. Solar hydrogen production by a two-step cycle based on mixed iron oxides. J Sol Energy Eng 2006;128:125–33.
- [6] Agrafiotis C, Roeb M, Konstandopoulos AG, Nalbandian L, Zaspalis VT, Sattler C, et al. Solar water splitting for hydrogen production with monolithic reactors. Sol Energy 2005;79:409–21.
- [7] Beghi GE. Development of thermochemical and hybrid processes for hydrogen production. Int J Hydrogen Energy 1985;10(7/8):431–8.
- [8] HYTHEC project. Available from: http://www.hythec.org.
- [9] Le Duigou A, Borgard J-M, Larousse B, Doizi D, Allen R, Ewan BC, et al. HYTHEC: an EC funded search for a long term massive hydrogen production route using solar and nuclear technologies. Int J Hydrogen Energy 2007;32:1516–29.
- [10] Jomard F, Feraud JP, Caire JP. Numerical modeling for preliminary design of the hydrogen production electrolyzer in the Westinghouse hybrid cycle. Int J Hydrogen Energy 2008;33:1142–52.
- [11] Dutta S, Morehouse JH, Khan JA. Technology assessment of advanced electrolytic hydrogen production. Int J Hydrogen Energy 1990;15:379–86.
- [12] Meteosat. Results of calculation.
- [13] Kiera M. Beschreibung und Handhabung des Programmsystems HFLCAL. Interatom Report; 1986.
- [14] Timmerhaus KD. Plant design and economics for chemical engineers. 4th ed. Chemical Engineering Series; 1991.
- [15] Kolb GJ, Jones SA, Donnelly MW, Gorman D, Thomas R, Davenport R, et al. Heliostat cost reduction study. Sandia Report Sand 2007-3293. Unlimited Release; Printed June 2007.
- [16] European concentrated solar thermal road-mapping. ECOSTAR Project SES6-CT-2003-502578. The German Aerospace Center; 2005.
- [17] Steinberg M, Cheng HS. Modern and prospective technologies for hydrogen production from fossil fuels. Int J Hydrogen Energy 1989;14(11):797–820.

- [18] U.S. Department of Energy. DOE H2A Analysis Database.
- [19] Air Liquide. Personal communication.
- [20] Plana-innova. Personal communication and request; 2007.
- [21] Alfa Aesar Johnson Matthey (Deutschland). Forschungschemikalien, Metall und Materialien, 2003-04 Allin-One Katalog, Stock No. 44120.
- [22] Alfa Aesar Johnson Matthey (Deutschland), Forschungschemikalien, Metall und Materialien, 2003-04 Allin-One Katalog, Stock No. 44375.
- [23] Degussa AG. Personal communication.
- [24] HYDRO, technical specification of electrolyzers. Available online, hydrogen technology products, <http://www.hydro. com/electrolysers/library/attachments/Brochures/49444_ ProductSheet_2.PDF>; 2007.
- [25] Dersch J, Schwarzbözl P. Dlr Cologne, Germany. Personal communication.
- [26] Mansilla C, Sigurvinsson J, Bontemps A, Maréchal A, Werkoff F. Heat management for hydrogen production by high temperature steam electrolysis. Energy 2007;32: 423–30.
- [27] Arbeitsgemeinschaft: Wuppertal Institut, DLR, ZSW, PIK; Erster Statusreport (CO₂-BMU_ZW_Bericht1.doc) September 2004; Forschungsvorhaben im Auftrag des Bundesministeriums für Umwelt, Naturschutz und Reaktorsicherheit; Ökologische Einordnung und strukturellökonomischer Vergleich regenerativer Energietechnologien mit anderen Optionen zum Klimaschutz, speziell der Rückhaltung und Speicherung von Kohlendioxid bei der Nutzung fossiler Primärenergien: <www.erneuerbareenergien.de/files/pdfs/allgemein/application/pdf/zb1_co2. pdf>.
- [28] Fahrni R. Hydrogen production an overview of hydrogen production methods and costs today. Term paper WS01/02 ETH Zürich. Institute of Energy Technologies; 2002.
- [29] Mueller-Langer F, Tzimas E, Kaltschmitt M, Peteves S. Techno-economic assessment of hydrogen production processes for the hydrogen economy for the short and medium term. Int J Hydrogen Energy 2007;32:3797–810.
- [30] HYDROSOL. http://160.40.10.25/hydrosol/hydrosol-home-en.html>.
- [31] HycycleS project. <www.HycycleS.eu>.