Evaluation of the Westinghouse cycles

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Abstract: - Hydrogen is industrially produced mainly from fossil fuels e.g. by natural gas steam reforming, coal gasification and as a by-product of naphtha reforming. A variety of technologies can be used for hydrogen production. In the long term, with respect of a lack of fossil fuels, hydrogen produced from water can play a very important role in the energy system. The goal of the water splitting thermo-chemical cycle is the replacement of thermal decomposition of water with several partial reactions. This paper deals with the one of most promising water splitting thermo-chemical cycle – the Westinghouse cycle. Simulation, optimization and overall evaluation (including calculation of efficiency and life cycle analysis) of the Westinghouse cycle were preformed and main obtained results are reported.

Key-Words: - hydrogen, hydrogen production, water splitting thermo-chemical cycles, Westinghouse cycle, life cycle analysis, solar energy

1 Introduction

Hydrogen is the most plentiful chemical element in the universe, but elemental hydrogen is relatively rare on Earth. Hydrogen is found f.e. in great abundance in stars and gas giant planets and plays a vital role in powering stars through nuclear fusion. Hydrogen can be produced from a variety of sources: such as fossil fuels, renewable sources or water with input from renewable energy sources (e.g. sunlight, wind, and hydro-power). A variety of technologies can be used, including chemical, biological, electrolytic, photolytic and thermo-chemical. Nowadays, hydrogen is industrially produced from fossil fuels (mainly from natural gas).

Hydrogen is considered to be an ideal energy carrier in the foreseeable future and it can be produced from water by using a variety of sources – solar energy, nuclear energy or fossil fuels.

Hydrogen may be produced from water using the process of electrolysis (at present only 4 % is produced by this process [6]). Moreover this process is presently more expensive commercially than production from natural gas.

Hydrogen production from water is the main goal of several research programmes.

Direct water dissociation is a non-practical way for obtaining hydrogen, due to relatively high temperatures (above 3 500 K) and the small content of hydrogen at the thermodynamic equilibrium. The aim is to perform hydrogen production by water thermolysis at reasonable temperatures (below 2 500 K).

It is possible to overcome aforementioned problems by using water splitting thermo-chemical cycles (WSTC), which are the processes for decomposition of water into hydrogen and oxygen via chemical reactions using intermediate substances which are recycled [1, 2, 4, 6].

One of the simplest and most promising water splitting thermo-chemical cycles was chosen for our study: the Westinghouse cycle. This cycle consists of the thermo-chemical and electrolytic steps [1, 4]. Detailed simulation of the cycles was performed in Aspen Plus code as well as optimization and overall evaluation.

The outline of this paper is following: Firstly is briefly described concept of water splitting thermo-chemical cycles. Basics approach of the Westinghouse cycle and description of the main sections of cycle follows in next section. Overall evaluation of the WH cycle is reported in the forth section. It contains optimization of SO_2/O_2 separation system, calculation of the efficiency (including general method to analyse thermodynamic efficiency) and life cycle analysis of the WH cycle. The main conclusions are summarized in the last section.

2 Water splitting thermo-chemical cycles

The water splitting thermo-chemical cycles (WSTC) are processes where water is decomposed into hydrogen and oxygen via chemical reactions using intermediate elements which are recycled. [1, 4, 6]

The concept of the WSTC was proposed in the 1960's and since then over one-hundred thermo-chemicals cycles were described.

The WSTC were extensively studied in the late 1970s and '80s, but had only a little interest in the past 10 years. While there is no question about the technical feasibility and the potential for high efficiency, cycles with proven low cost and high efficiency have yet to be developed commercially [9]. The WSTC represent alternative way of hydrogen production without using fossil fuels. Several of them have been successfully tested and evaluated including their chemistry, bench scale studies and process engineering studies.

Some of the WSTC are purely chemical processes and others contain also electrochemical steps and consist of both endothermic and exothermic reactions.

The main endothermic reactions take place at temperature running in the range 700 – 1200 °C. Therefore, only the high temperature sources could be chosen for this process e.g. solar or nuclear energy.

Basic idea of the WSTC which use solar energy (Fig. 1) is to concentrate the sunlight with the help of solar systems and to obtain the heat at high temperature for driving a chemical transformation and production of storable and transportable fuel [3]. The products at high temperature exiting solar reactor are separated and quenched. Finally an ideal fuel cell is used to produce the work and the reactants are sent back to the solar reactor.

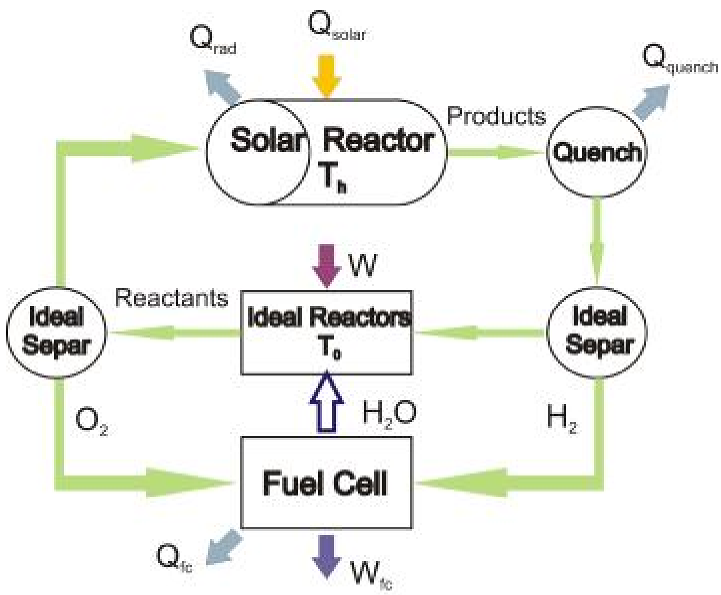

Fig. 1 Schematic model of solar energy conversion

3 Basics of the Westinghouse cycle

The Westinghouse cycle (WH cycle) is a two-step thermo-chemical cycle for decomposition water into hydrogen H_2 and oxygen O_2.

Hydrogen is produced by electrolysis. Sulphur dioxide SO_2 and water H_2O are reacted electrolyticaly to produce hydrogen H_2 and sulphuric acid H_2SO_4. The resultant sulphuric acid H_2SO_4 is vaporised to produce steam and sulphur trioxide SO_3, with the latter being subsequently decomposed at high temperature into sulphur dioxide SO_2 and oxygen O_2.

The oxygen is available as a process by-product. The required thermal and electrical energy can be provided by concentrated sunlight to reach higher temperature [4]. The reactions in the WH cycle are following:

$$H_2SO_4\,(g) = H_2O(g) + SO_2(g) + \tfrac{1}{2}\,O_2\,(g) \quad (1)$$
thermo-chemical, 800 - 850 °C

$$SO_2\,(g) + 2\,H_2O\,(l) = H_2(g) + H_2SO_4\,(l) \quad (2)$$
electrolysis, 25 - 100 °C

The Westinghouse cycle is schematically illustrated in Fig. 2. There are four major sub–systems in the cycle: ***concentrator, decomposer, separator and electrolyser***.

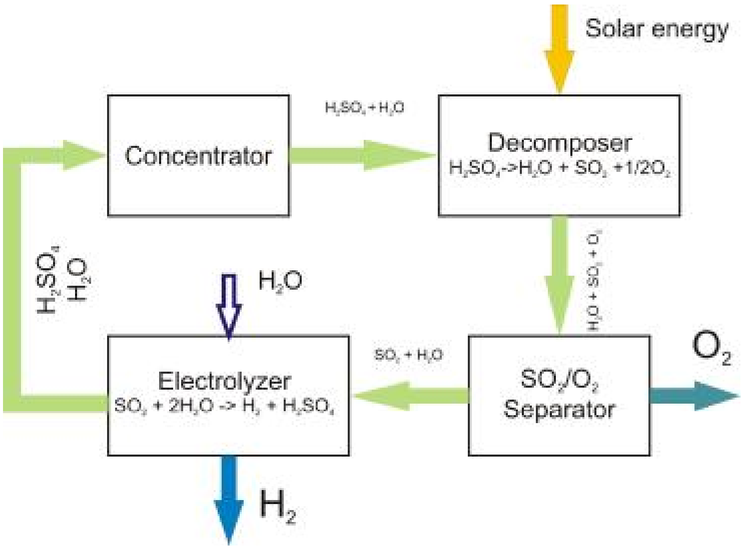

Fig. 2 Westinghouse cycle – simplified model

Following short description of four aforementioned major sub–systems of the WH cycle:

- ***Concentrator***: The role of the concentrator is to remove water from sulphuric acid by heating and flashing [1, 4]. They can be separated due to different boiling points. The efficient liquid mixture of sulphuric acid and water (in our case approx. 40 % H_2SO_4 and 60 % H_2O) is sent to the decomposer and vaporized water to the electrolyser.

- ***Decomposer***: According to the obtained results from [1, 4], operation conditions for solar reactor corresponding to pressure of 1 bar and temperature of 830 °C were set. The reaction is endothermic and the high temperature is required for the sulphuric acid decomposition. Therefore, only the high temperature heat sources are usable for this process (solar or nuclear energy). In decomposer sulphuric acid H_2SO_4 is decomposed into sulphur trioxide SO_3, which is latter being decomposed at high temperature into sulphur dioxide SO_2 and oxygen O_2. The hot decomposed gas is sent to the cooler and then to the separator tank where vapour mixture of SO_3, SO_2 and O_2 is separated. Vapour mixture of SO_2 and O_2 is transmitted to the separator sub-system and liquefied SO_3 to the electrolyser.

- ***Separator:*** Vapour mixture of SO_2 and O_2 is compressed by a compressor (to achieve high pressure for efficient separation) and then is sent to the separation tank. A large fraction of liquid SO_2 is transferred to the heater and then to the electrolyser. Gas O_2 and portion part of SO_2 is transferred to chiller for future separation which nearly completes separation of SO_2 from O_2 at a very low temperature. The separated portion part of SO_2 is sent to the electrolyser and O_2 as a by-product can be stored for future utilization. This two steps separation permits to obtain a very pure oxygen at the inlet as the by-product. SO_2/O_2 separation sub-system was optimized to maximize O_2 production in gas phase and SO_2 production in liquid phase. The maximization of SO_2 has impact to the hydrogen production.

- ***Electrolyser:*** The role of the electrolyser is to produce hydrogen at the cathode and sulphuric acid at the anode. Sulphuric acid is then circulated through a closed loop. [4]

4 Evaluation of the Westinghouse cycle

Industrial scale-up studies have a great importance, for the assessment of safety aspects of the process, the feasibility of the main components at industrial scale, and H_2 production costs. [6]

In our study, the improvement of the Westinghouse cycle was focusing on optimization of SO_2/O_2 separation sub-system because maximization of SO_2 entering to electrolyzer has the big impact to the hydrogen production and therefore also for efficiency of the cycle. Efficiency of cycle with respect to its thermodynamic theoretical values was calculated and the main results are reported.

4.1 Simulation and optimization of the Westinghouse cycle

Aspen Plus (Aspen Plus®, Aspen Technology, Inc. (AspenTech.)) was chosen as the process simulator for this work. Aspen Plus® is employed for chemical process simulation and for developing process flow sheet, process analyses and optimization. It includes the capability of simultaneously regressing model parameters of many different types in order to generate a thermodynamic model for the specific chemical system.

There are many different modelling techniques. The Peng-Robinson method using Peng-Robinson equation of state was chosen for the WH cycle simulation, following the recommendations in [3].
Complete simulation model is illustrated in Figure 5.

SO_2/O_2 separation sub-system was optimized to maximize O_2 production in gas phase and SO_2 production in liquid phase. The maximization of SO_2 has the significant impact to the hydrogen production. The SO_2/O_2 separation section is shown in Figure 3.

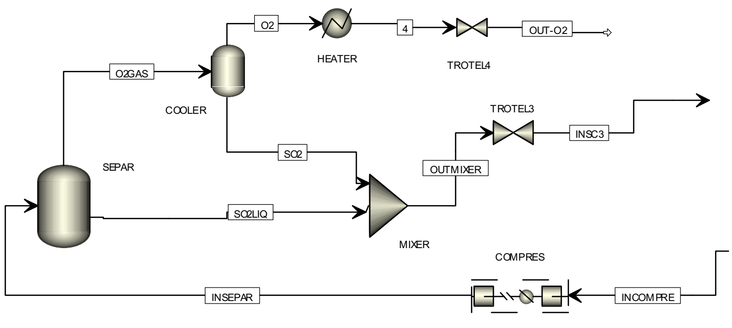

Fig. 3 SO_2/O_2 separation – optimized sub-system of the WH cycle

Vapour mixture of SO_2 and O_2 is compressed by the compressor to achieve high pressure (20 bar) for efficient separation and then is transmitted to the separation tank. A large fraction of liquid SO_2 is sent to mixer, later is heated and then sent to the electrolyser. The mixture of O_2/SO_2 in gas phase is transported to the cooler for follow-up separation which nearly complete separation of SO_2 from O_2 at very low temperature (- 45 °C). The separated portion part of SO_2 is also send to mixer and after heating to the electrolyser. Two steps separation allows obtaining very pure oxygen at the inlet as the by-product which could be, for example, stored for future utilization.

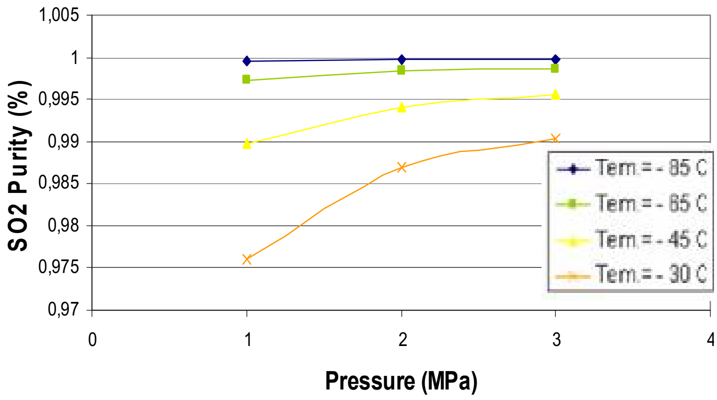

Fig. 4 Comparison of SO_2 purity at different conditions at the outlet of SO_2/O_2 separation system

The mass fraction and the purity of SO_2 outlet entering the electrolyzer were calculated at different conditions (Fig. 4 and Fig. 6). Using sensitivity analysis, the best conditions to optimize the hydrogen production were chosen. The sensitivity analysis was made by different conditions for cooler: four different temperatures (- 85 °C, - 65 °C, - 45 °C, - 30 °C) and three different pressures (10 bar, 20 bar, 30 bar).

Mass fraction was calculated as:

$$MFrac_{so_2} = \frac{MF_{so_2}}{TMF} \qquad (3)$$

Where
MFrac $_{SO2}$ – Mass Fraction of SO_2 (%)
MF_{SO2} – Mass flow of SO_2 (inlet to electrolyser, $kg.s^{-1}$)
TMF – Total mass flow (inlet to electrolyser, $kg.s^{-1}$)

Purity of SO_2 was calculated as:

$$P_{so_2} = \frac{MF_{SO_2}}{MF} \qquad (4)$$

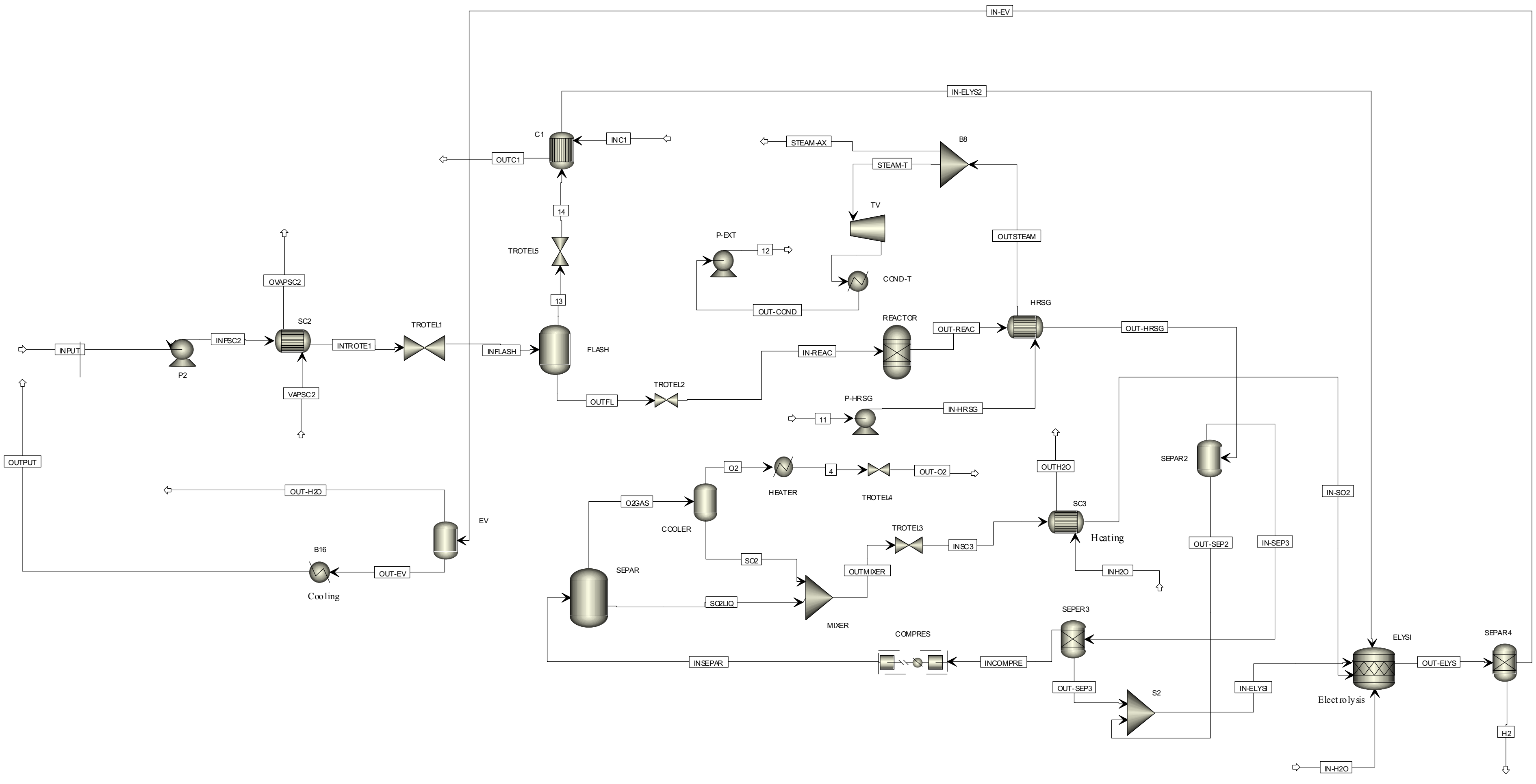

Fig. 5 Simulation of Westinghouse cycle in AspenPlus

Where
PSO2 - Purity of SO_2 (%)
MF -Mass flow of SO_2+O_2 (inlet to electrolyser, kg.s^{-1})

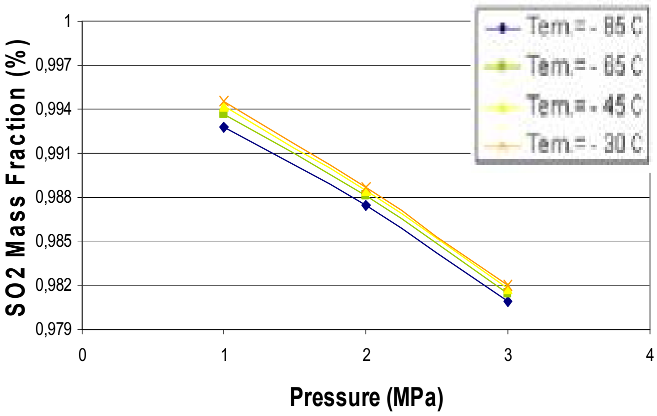

Fig. 6 Comparison of SO_2 mass fraction at different conditions at the outlet of SO_2/O_2 separation system

The best results were obtained at temperature T=-85 °C and pressure p = 30 bar. The energy needed to achieve these conditions has impact to overall efficiency of the thermo-chemical cycle and a good compromise in the operating conditions is obtained for T = - 45 °C and pressure p = 20 bar. The final mole fraction at the outlet from SO_2/O_2 separation system is 98,8 % and the purity of recycled sulphur dioxide is 99,4 %.

4.2 Efficiency of the Westinghouse cycle

The theoretical maximum efficiency of the energy-conversion processes is limited by the Carnot efficiency of an equivalent heat engine.

The cycle efficiency based on the 1st thermodynamic law of the closed-cycle is calculated as:

$$\eta_c = \frac{W_{net}}{Q_{solar}} \qquad (5)$$

Where W_{net} [kJ/mole] is the maximum useful net work that the leaving products from the reactor can produce in the case they are combined at temperature T (the temperature of reactants entered to solar reactor) and a total pressure of 1 bar [3]. Q_{solar} [kJ/mole] is total solar heat input.

The Carnot efficiency η_{car} could be expressed:

$$\eta_{car} = \frac{W_{\max}}{Q_{solar}} = 1 - \frac{T_0}{T_h} \qquad (6)$$

Products exiting the solar reactor at T_h are cooled rapidly to T_0.
The maximum available work W_{max} [kJ/mole] can be calculated as the sum of the net useful work plus the lost work due to irreversibilities in the solar reactor and during quenching.
Irreversibilites in the solar reactor $Ir_{reactor}$ [kJ/mole/K] arise from the non-reversible chemical transformation and reradiation losses to the surroundings at temperature $T_{0.}$ Ir_{quench} [kJ/mole/K] represents the quenching irreversibility.

$$W_{\max} = W_{net} + T_0(Ir_{reactor} + Ir_{quench}) \qquad (7)$$

The results are summarized in table 1.

Efficiency of the WH cycle was calculated as ratio between net useful work with respect to solar energy in solar reactor to decomposed H_2SO_4. Results were compared with the theoretical values (Table 1).

The WH cycle total work calculation consider following:

- mass flow rate of hydrogen M_{H2} and the low caloric value of hydrogen LCV_{H2},
- amount of work produced in turbine W_{Turb} during the cycle,
- work needs in the electrolyser W_{EL}, compressor W_{Com}, cooler W_{Cool} and pumps W_P.
- Low caloric value of hydrogen is 120 MJ/kg

Products leaving reactor at very high temperature (1103 K) are cooled down. Water used for their cooling in heat recover steam generator is lately used in turbine to produce the work and a part of this water is used as an auxiliary steam in the others pars of the cycle.

Therefore the WH cycle efficiency can be calculated following:

$$\eta_{eff} = \frac{(M_{H2} LCV_{H2} + W_{Turb}) - (W_P + W_{EL} + W_{Com} + W_{Cool})}{Q_{Solar}} \quad (8)$$

Table 1: Simplified results of cycle efficiency

	Unit	WH cycle
$M_{H2produced}$	kg/hr	378
W_{net}	kJ/mole	158
Q_{solar}	kJ/mole	479
W_{turb}	kW	809
$W_{P,EL,Com,Cool}$	kW	5411
Total work	kW	8024
Solar power	kW	26957
η_{car}	%	64
η_c	%	33
η_{eff}	%	30

4.3 Life cycle analysis of the Westinghouse cycle

The evaluation process of life cycle analysis (LCA) covers the whole life cycle (considering three main phases ***construction, operation*** and ***dismantling***), including extraction of raw materials, fabrication processes, transport, distribution, utilization, production, re-use, internal recycle and final disposal.

The goal of LCA is to compare the environmental performance of products, to be able to choose the least burdensome one. The concept can be used to optimize the environmental performance of a single product. [14, 15, 16, 17]

In methodology the raw materials are considered from their birth through their evolution and transformation and are followed before and during the production process up to the final disposal of the reject materials.

Moreover the energy spent to achieve these processes is also considered, and the energy data is related to all the impacts concerning to the production. Using this analysis also the secondary processes related to the main process are taken into account. [15, 17]

For investigation of hydrogen production impacts from the WH cycle was used LCA tools.

The inlet and the outlet streams for process simulation consist of the raw material flows and waste/emission flows of operation phase. The simplified model for operation phase of the WH cycle is schematically shown in Fig. 7.

Fig. 7 Simplified models of the WH cycle to perform LCA

4.3.1 Simulation in SimaPro 7.0

Simulations were performed in SimaPro 7.0, which is basically a database able to reconstruct the "history" of several processes and materials and to aggregate the elemental pollutants inventory in order to obtain values for the selected environmental effect indicators. [16]

A critical issue is the definition of the functional unit because all measurement will be referred to it during LCA. The functional unit states is reference unit to which all inlet and outlet flows will be referred. For our case as ***a functional unit*** was chosen ***1 kg of produced hydrogen***.

The choice of using very simplified model (Fig. 7) of the power plant was performed because all other flows in the system are internal and therefore have no impacts on environment.

Oxygen was considered as an avoided product of the cycle because it is possible to reuse it e.g. in fuel cells.

The inputs parameters for operation phase were obtained from AspenPlus simulation and are mentioned in section 4.1 and 4.2 of this paper. They are summarized in table 2.

Some inputs parameters for construction phase of the WH cycle were established following:

- Nominal power of plant – 26 MW
- Plant life – 20 years
- Working hours – 2600 hours/year
- Effective surface of plant – 26000 m^2
- Number of mirrors – 376

Table 2 Data for the Westinghouse cycle used in SimaPro - System process analysis

OUTPUT		Unit
H_2	1	kg
O_2 in the air	7,61	kg
SO_2 in the air	0,19	kg
INPUT		Unit
Water	9,17	kg
H_2SO_4	0,19	kg
Electric power	12,86	kW

All the data are related to 1 kg of produced hydrogen what is the functional unit to which LCA is referred.

Different environmental issues can be much different concern to the local audit, depending on traditions, climate conditions, etc. However, there are well-established rules and trends which have determined the development of several largely accepted assessment methods (e.g. EPS, EDIP, Eco-Indicator 95, Eco-Indicator 99). There are large differences among methods for definition of categories (otherwise they consider different amount of impact categories). [10, 11]

In our simulation was chosen ***Eco-Indicator 99*** which is a "Damage-oriented" method of LCA. All types of impact are reduced to three damage macro-categories, which are originated by the original impact categories. Eco-Indicator 99 refers to three impact categories: ***human health, ecosystem quality and resources.*** [14, 15, 16]

4.3.2 Results of the simulation

The networks show only a part of the sub-processes considered in the overall processes, due to the different impact linked to each sub-process. In fact in SimaPro it's possible to choose the node cut-off (the % of impact related to a process that permits this process to be taken into account).

Normalized results of life cycle analysis for hydrogen production via Westinghouse cycle are shown in Fig. 8.

On the y-axis is shown the impacts referred to equivalent inhabitants. On the x-axis are the impact categories according to the selected method Eco-indicator 99.

The inventory results contain hundreds of different emissions and resource extraction parameters. Once the categories are defined, each emission must be converted into the designated category indicator.

Normalization is a procedure needed to show to what extent an impact category has a significant contribution to the overall environmental problem. This is done by dividing the impact category indicators by a "normal" value. There are different ways to determine the "normal" value. The most common procedure is to determine the impact category indicators for a region during a year and, if desired, divide this result by the number of inhabitants in that area.

The normalization is referred to annual emission of inhabitant in areas like Europe, for instance the environmental effects that an average European person causes in one year.

After this step a chart is obtained comparing different system on the base of inhabitant equivalents, obviously scores for ozone layer depletion, eutrophication, pesticides and carcinogens are very low in absolute terms.

Normalization reveals which effects are large and which are small in relative terms. However it does not yet say anything about relative importance of the effects. In fact a small effect could be the most important so the weighting step would be necessary. [14, 15]

Oxygen is considered as an avoided product therefore in y-axis has negative value. There is visible higher impact of respiratory inorganic and fossil fuels compounds on account of electric power needed in some part of the cycle.

Comparison of LCA of the WH cycle with other processes for hydrogen production (e.g. pyrolysis, gasification) will be performed in the future work for better understanding of process impact.

5 Conclusion

Detailed simulation of the Westinghouse cycle was performed in AspenPlus code. The goal of the optimization process of the WH cycle was to improve the conditions in separation section. Purity of O_2 at the outlet was increased during the optimization process and amount of recycled SO_2 was maximized. The maximization of SO_2 has significant impact to the hydrogen production because SO_2 and water H_2O are reacted electrolyticaly in electrolyser to produce hydrogen H_2 and H_2SO_4 which is later recycled in the process.

Efficiency of the Westinghouse cycle was calculated with respect to thermodynamic theoretical values.

One of disadvantages of the WH cycle is electrolysis step and the high energy consumption needs for SO_2/O_2 separation process.

The thermo chemical water splitting cycles which use solar energy as a heat source are very promising due to utilization of renewable sources for hydrogen production and quit high efficiency of the cycle.

Some results of LCA of the WH cycle are reported. Dismantling phase will be considered in the future in order to realize a complete LCA study. Comparison of LCA of the WH cycle with other processes for hydrogen production will be performed in future work.

There are still needs of improvement of the Westinghouse cycles and cumulation of results. It is also advisable to compare obtained results with experimental results and future improvement of simulations following the experiment. Future improvements could enhance efficiency of the WH cycle and maximize hydrogen production.

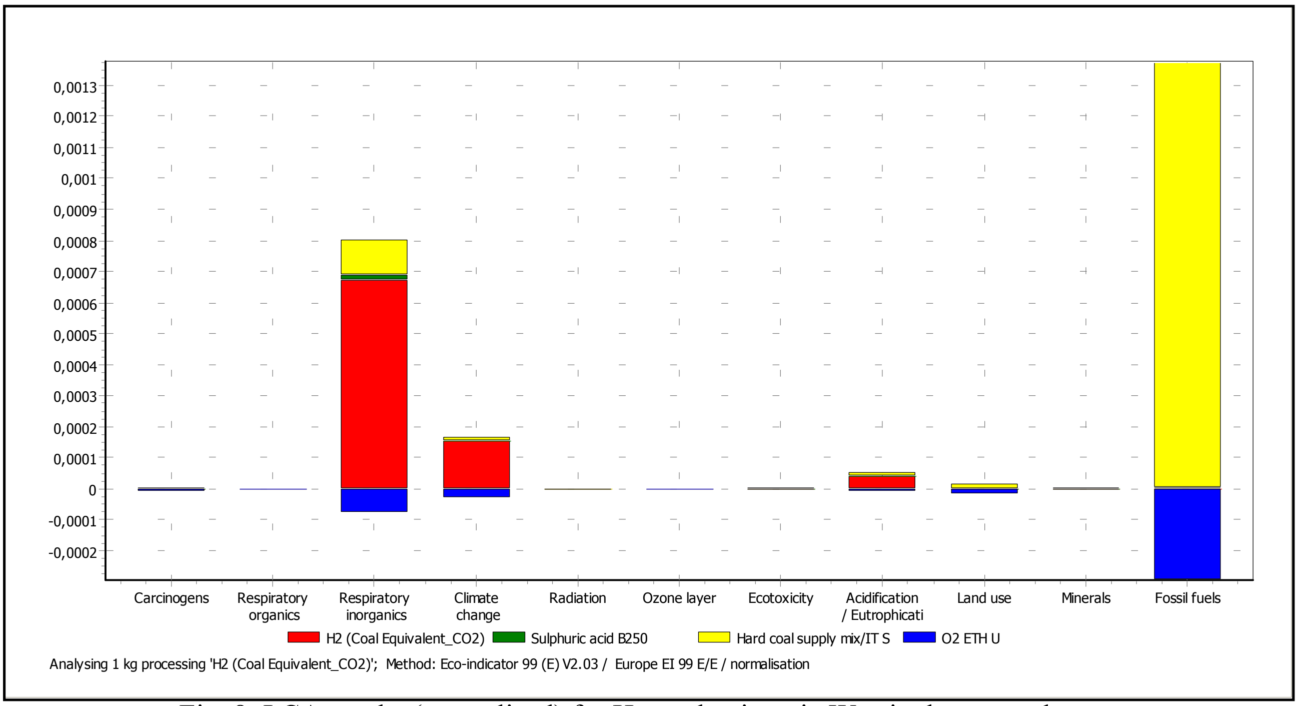

Fig. 8 LCA results (normalized) for H_2 production via Westinghouse cycle

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