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Techno-economic analysis of H₂ production processes using fluidized bed heat exchangers with steam reforming – Part 1: Oxygen carrier aided combustion

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Abstract

This work presents the techno-economic assessment for a new process where a fluidized bed heat exchanger (FBHE) is used as heat source for steam reforming in a hydrogen production plant. This suggested process configuration is compared with a reference case representing a conventional steam methane reforming (SMR) large-scale hydrogen production plant. The use of a FBHE as a heat source for the endothermic reforming is an advantage because of the high heat transfer coefficient to the reformer tubes. The suggested process configuration utilizes oxygen carrier particles as bed material and a bubbling fluidized bed reactor with immersed reformer tubes to ensure sufficient heat production for the reforming and improved heat transfer to the reformer tubes compared a conventional plant. The results include a comparison of hydrogen production efficiency and levelized production costs (LCOH) of the two plants where the production efficiency is more than 11% higher and the LCOH is more than 7% lower for the suggested process configuration.

Highlights

- Techno-economic assessment of hydrogen production based on Aspen Plus simulation.
- SMR plant with a fluidized bed heat exchanger was compared to a conventional plant.
- The H₂ production efficiency is improved by 11.6% compared to the conventional plant.
- The levelized production cost is 7.4% lower for the new plant configuration.

Keywords

hydrogen; steam reforming; oxygen carrier aided combustion; fluidized bed heat exchanger;

Abbreviations

AFR	Air-to-fuel ratio
BEC	Bare Erected Cost
BUA	Bottom-up approach
C&OC	Contingency and Owner's Cost
CAGR	Compound annual growth rate
C28	Mn-based perovskite oxygen carrier material, $\text{CaMn}_{0.775}\text{Mg}_{0.1}\text{Ti}_{0.125}\text{O}_{3-\delta}$
CCF	Capital charge factor
CEPCI	Chemical Engineering Plant Cost Index
CLC	Chemical-looping combustion
COND	Condenser
EBTF	European Benchmarking Task Force
ECO	Economizer
EPC	Engineering procurement and construction
EVA	Evaporator
FBHE	Fluidized bed heat exchanger
Fuel gas	Gas stream consisting of PSA off-gas and supplementary fuel
GFF	Gas-fired furnace (in conventional SMR)
HEN	Heat exchanger network
HP	High pressure
HRSG	Heat recovery steam generator
HT	High temperature
HX	Heat exchanger
IP	Intermediate pressure
LCOH	Levelized cost of hydrogen
LHV	Lower heating value
LP	Low pressure
LT	Low temperature
NETL	National energy technology laboratory
NG	Natural gas
NG Feed	Natural gas fed to the reformer tubes
NG fuel	Natural gas used as supplementary fuel
NGCC	Natural gas combined cycle
O&M	Operations and maintenance
OC	Owner's cost
OCAC	Oxygen carrier aided combustion
Off-gas	Low pressure stream of gases obtained from the PSA
PSA	Pressure swing adsorption
S/C ratio	Steam-to-carbon ratio (molar basis)
SH	Superheater
SMR	Steam methane reforming
ST	Steam turbine
SUCCESS	Research project aimed for scale-up of CLC
TIC	Total installation cost
TPC	Total plant cost
TDPC	Total plant direct cost
WGS	Water-gas shift

Symbols

A_{req}	Area required to transfer the heat for steam reforming to the reformer tube
C	Equipment cost with the capacity S
C_o	Reference cost of equipment with reference capacity S_o
c_p	Heat capacity (J/K)
d_{cat}	Equivalent diameter of catalyst pellet
d_t	Reformer tube diameter
dT_{min}	Minimum temperature difference
E	Emissions of CO_2 from hydrogen production plant
ε	Voidage (void fraction)
e	Emissivity
σ	Stefan-Boltzmann's constant, 5.67×10^{-8} (W/(m ² K ⁴))
g	Gravitational constant, 9.81 (m/s ²)
G	Superficial mass velocity (kg/(m ² s))
$h_{o,\text{FBHE}}$	Bed-to-tube heat transfer coefficient (W/(mK))
k	Thermal conductivity (W/(mK))

L_t	Reformer tube length (m)
μ	Dynamic viscosity (kg/(ms))
m	Number of installed units (used in cost function)
$\dot{m}$	Mass flow (kg/s)
$\dot{M}$	Molar flow (mol/s)
N_t	Number of reformer tubes
η	Efficiency (%)
ξ_1, ξ_2	Correction factor for tube bundle
ρ	Density (kg/m ³)
Pr	Prandtl number, $\frac{c_{p,g}\mu_g}{k_g}$
Q_{th}	Heat delivered from steam export (W)
Re	Reynolds number
S	Capacity (scaling parameter in cost functions)
ϕ	Sphericity
u_o	Superficial gas velocity (m/s)
U_o	Overall heat transfer coefficient (W/(m ² K))
$\dot{V}_{H_2}$	Volumetric flow per hour of produced hydrogen (m_{n,H_2}^3 /hr)
W	Power (W)
T	Temperature (°C)
x	Year when reference equipment cost was estimated
Y_{H_2}	Hydrogen yield

Indices/Exponents/subscripts

'	Case neglecting use of LP steam as heat export
APH	Preheated air entering the reformer furnace
b	Bed
δ	Degree of oxygen deficiency
el	Electricity
eq	Equivalent
exp	Export
fixed	Fixed bed
g	Gas
i	Inside
liq	Liquid
LM	Logarithmic mean
n	exponent in scaling cost function
o	Outside (reformer tubes) or superficial (for velocity)
O&M, fix	Fixed operations & maintenance costs
O&M, var	Variable operations & maintenance costs
sat	Saturated
p	Particle
th	Thermal
tot	Total
reference	Reference plant for heat or electricity production
reform	Reformer, reformer gas
vap	Vapor
w	Wall

1 Introduction

The annual hydrogen production corresponds to approximately 2% of the primary energy demand [1, 2]. Hydrogen is mainly used in the refining, chemical and industrial gas industries with several end-use applications. Hydrogen can largely be transformed to any form of energy, and is thus highly applicable for multitude of applications [3]. Refineries are estimated to account for 65% of the hydrogen production capacity and together with ammonia and methanol production plants these plants constitute 92% of the production capacity [4]. Hydrogen could play an important role in the future energy system. The Compound Annual Growth rate (CAGR) for hydrogen is expected to exceed 6% from 2016 to 2022 [5] which is mainly driven by legislation for regulating desulfurization of petroleum products and increased demand for hydrogen in the transportation sector. The recent initiative HYBRIT to replace coal with hydrogen in the steel-making process is another example of a potential source for increased demand for hydrogen in the future [6].

Approximately 95% of the hydrogen produced today is based on fossil fuels [1] and hydrogen production results in more than 1% of the global CO₂ emissions [3] corresponding to approximately 500 Mton CO₂ per year. Hydrogen production based on fossil fuels is considered to be the most economically feasible alternative during the next 20 years and these fuels could also play an important role in the longer term (20-50 years) [7]. The two main production routes are steam reforming of methane/natural gas (SMR) (48% of produced hydrogen) and reforming of oil/naphtha (30%) [8] and the rest is produced from coal gasification (18%) and electrolysis (4%) [3]. Large-scale SMR production is considered to be the benchmark technology for hydrogen production today based on its general economic feasibility globally [3].

The conventional steam reforming process is based on catalytic reforming of natural gas at high temperature (800-950°C) and high pressure (15-40 bar). The endothermic reaction (Eq.(1)) takes place in high-alloy reformer tubes where a catalyst (typically nickel [9]) is placed on a pellets support material [10]. The steam-to-carbon (S/C) ratios are commonly 2-6. The tubes are heated by external gas burners placed in a furnace where different tube and burner arrangements can be used [10].

The reformer is followed by one or more water-gas-shift (WGS) reactors to adjust the H₂/CO-ratio which in ammonia and hydrogen production plants should be as high as possible [10]. The CO is converted according to the equilibrium-limited and slightly exothermic WGS reaction (Eq.(2)), which is favoured by low temperatures.

The products from reforming and shift reactors are cooled to close to ambient temperature and sent to a pressure swing absorber (PSA) where a high purity hydrogen stream is obtained (up to 99.999% purity depending on the market requirement [11]). A low pressure off-gas stream is generated which is rich in CO₂, but also combustible gases in form of CO, CH₄ and H₂. The off-gas is typically fed as fuel in the reformer furnace [11], as is additional fuel in form of natural gas (refinery gas could also be used). Combustion of off-gas and natural gas provides the heat for the endothermic reaction (1).

SMR plants typically generate significant amounts of excess heat, which in general should be avoided [10]. One of the main reasons for the excess heat generation is the low heat transfer coefficients to the outside of the reformer tube wall, where the driving force is mainly radiation from combustion at high temperatures [12]. The high temperatures required also results in thermal NO_x emissions. Further, the heat transfer to the tube wall from the outside is typically the rate limiting step for reaction (1). Thus, if the heat transfer to the tube wall could be improved, a lower furnace temperature could be used to: i) reduce the fuel consumption of

supplementary fuel, ii) reduce the amount of excess heat generation, iii) reduce thermal NO_x emissions, iv) reduce the risk of temperature hotspots on the tube surface (a common reason for reformer tube failure) which is caused by uneven heat flux to the reformer tubes which is difficult to control by flame combustion.

Fluidized bed technology is used in a wide range of applications such as fluid catalytic cracking (FCC), roasting of ores and various drying processes where one of the key benefits is the outstanding heat and mass transfer characteristics of the fluidized bed. One example of an application where very high heat transfer coefficients to tubes are observed is fluidized bed heat exchangers (FBHEs). A water/steam-mixture is here commonly present in the immersed tubes which are used to remove heat. This kind of design has been used for various exothermic chemical processes such as production of acrylonitrile from propylene [13] (Sohio process) and production of maleic anhydride from benzene [13]. Another example of use of external FBHE-units in circulating fluidized bed (CFB) boilers [14] which can replace heat exchange surfaces otherwise placed in the furnace, where lower heat transfer coefficients are observed [14, 15]. The FBHE is placed after the cyclone solids outlet, where fluidization can be done at low superficial gas velocities, around 0.2-0.5 m/s to avoid tube erosion. The heat transfer coefficient is also considered to be rather uniform in a bubbling fluidized bed which could possibly result in a reduction of local hotspots on the tubes.

Despite the successful implementations of FBHE for heat extraction at high temperature levels, to the author's knowledge there are currently no examples of implementations of FBHEs as a heat source for tubular chemical reactors. The idea to immerse steam reforming tubes in bubbling fluidized beds was first presented by Rydén et al. [16]. For a steady-state process a circulating fluidized bed system would be required, where hot particles continuously enter the FBHE, transfer the heat to the tube filled with catalyst where the reforming is occurring and exit the reactor at lower temperature. Alternatively, a fluidized bed combustor could be considered where natural gas and air/oxygen reacts and the heat is transferred to the immersed tubes. However, it has been observed that, in bubbling fluidized bed systems fluidized with air and methane/natural gas with silica sand bed material at moderate temperatures (below 850°C), most of the methane is converted in the freeboard [17, 18]. Heat generated in the freeboard would be of little use in a FBHE since the reformer tubes should be placed in the dense bed, where the heat transfer coefficients are significantly higher. Hence a system where the heat can be generated in the bed would be of high interest.

Chemical looping systems are examples of fluidized bed systems where complete conversion of methane/natural gas has been observed for multiple different oxygen carriers, even without presence of air and at temperatures similar to the target temperature for this application [19-25]. Recent review articles provides an overview of the chemical looping research area [26-29]. Oxygen carriers could also be used in a single fluidized bed system which is the idea behind oxygen carrier aided combustion (OCAC) where oxygen carriers are used as bed material instead of sand [30]. The oxygen carrier introduce new mechanisms for conversion of fuel, transport of oxygen in space and time [30] and stable fuel molecules such as methane can be converted through a heterogeneous reaction with the oxygen carrier without the need for gaseous oxygen. The concept of OCAC has been tested in industrial sized units with various fuels [30, 31].

The aim in this work is to integrate SMR with a single FBHE heated with the OCAC process without altering the traditional layout of an SMR plant except for the steam reformer furnace. This would present several possible benefits from both a thermodynamic and economic point of view.

The main objective of this paper is to perform a techno-economic assessment of a large-scale plant for hydrogen production using a single FBHE as a heat source for steam reforming of natural gas. The analysis covers both a thermodynamic as well as an economic analysis, including both capital costs and operational costs. The suggested process was recently introduced in an article by Stenberg et al. [32], where a more basic thermodynamic analysis was made displaying possible benefits compared to the conventional SMR process. The analysis is based on a more detailed Aspen Plus simulation model including all major components in the plant and thermal integration. The stream data obtained from this model is then used as starting point for reactor design, thermodynamic performance evaluation and economic assessment. Although significant work which has been published covering conventional SMR in general as well as techno economic assessments of large-scale plants [33-36], a model of a reference SMR plant is provided in this work to present a fair comparison, in terms of process performance and profitability. An overview of possible methods for industrial hydrogen production can be found in recent review articles [37-41].

2 Plant description

Two different plants for hydrogen production are modelled in Aspen Plus to assess the performance of the plants in terms of energy consumption, CO₂ emissions and hydrogen production cost. Both plants are dimensioned for large scale hydrogen production of 100000 m_n³/h (2.50 kg/s) pure hydrogen. The assumed hydrogen production plant has a size common for hydrogen production plants installed in refineries today. The plant in this study is assumed to be built on the west coast of Sweden. The plant design is a greenfield design with no restrictions in the plant area and no constraints in the delivery of equipment etc. The pressure of the delivered hydrogen is assumed to be 70 bar based on the maximum pressure of hydrogen gas pipelines [42] and similar to the pressure used at Preemraff Lysekil [43], which is an existing refinery on the west coast of Sweden. The plant is assumed to be delivering hydrogen to industrial consumers in the close vicinity of the plant.

2.1 Conventional steam reforming plant – Case A

The state-of-the-art steam reforming plant for hydrogen production is to a large extent based on the plant described by Martínez et al. [34]. The reference SMR plant is hereafter also referred to as case A. In this plant (see Figure 1), the natural gas stream delivered at elevated pressure is first purified in a hydrodesulfurization step where sulfur is removed. Prior to the desulfurization step (DES), the NG is mixed with a recycle stream of H₂ and pre-heated to around 370°C to convert organic sulfur compounds to H₂S in the DES step by using a CoMo catalyst on porous alumina support [10]. The produced H₂S is then adsorbed in a ZnO fixed bed reactor. The DES step is however not necessary for low-sulfur fuels [44]. The desulfurized gas is mixed with steam to achieve a steam-to-carbon molar ratio of 2.7 to avoid carbon deposition, to ensure a high steam-to-dry gas ratio in the WGS reactors and preserve the Fe-based catalyst [34]. The steam-to-carbon ratio is to a large extent limited by material constraints and it is possible to operate with even lower S/C ratios of 1.8-2.5 in modern SMR plants [10], although 2.7 is a commonly assumed value [33, 34, 45].

After desulphurization, the gas mixture of NG and steam is fed to a pre-reformer with a nickel catalyst at 500°C, where higher hydrocarbons are converted to methane and carbon dioxide. Here, the reforming and shift reactions reach equilibrium, resulting in some CH₄ conversion [10]. The use of a pre-reformer can reduce the heat duty in the reformer by 5-10% [46] and it also reduces the risk of carbon formation by decreasing the hydrocarbon concentration.

The gases are preheated to 620°C before the steam reformer. The reformer tubes are heated in a gas-fired furnace (GFF) to a temperature of 900°C for the produced syngas and approximately 80% of the methane is converted according to Eq.(1). For this plant a reforming pressure of around 32 bar was selected, which is typical for industrial applications. The produced syngas is then cooled down to 330°C. The system is integrated with a steam cycle in the heat exchanger network (HEN). In addition to producing HP steam at 92 bar, 485C, steam from this network is used for the reformer, delivered as intermediate pressure steam to the pre-reformer. The hot gases from the reformer are also to preheat the feed streams to the furnace and reformer. See Figure 1 for the system flowsheet.

To avoid metal dusting (see [47] for more information) in the high temperature heat exchangers following the reforming unit, part of the evaporator load was added before the superheaters to provide better gas cooling (see Figure 6). The cooled syngas is fed to a HT shift reactor (commonly carried out with a Fe-based catalyst) where approximately 70% of the CO is converted according to Eq.(2)). This plant has only a HT WGS reactor while LT WGS is normally present in plants for ammonia production or in cases where CO₂ sequestration is included to enhance H₂ yield even more and obtain a higher CO₂ concentration [33, 34, 45]. The gases are then cooled to ambient temperature and the condensed steam is removed before the PSA. The PSA has a H₂ separation efficiency of 90% [34] and a purity of 100%. The PSA off-gas is delivered at 1.1-1.7 atm typically [7] and is mixed with additional untreated NG, preheated and then fed to the reformer furnace where it provides heat for the steam reforming. The required outlet temperature of the reformer furnace is calculated based on a model of the heat transfer in the reformer taking the radiation into account (see below). The hot flue gas is used to heat the mentioned cold process streams, preheating of feedwater and additional HP steam production. An IP steam turbine generates electricity and slightly superheated LP steam at 6 bar. The LP steam could be used for various purposes depending on the plant at hand. The minimum stack temperature for the flue gas is set to 150°C which is adopted at the SMR plant at Preemraff Lysekil. At Preemraff Lysekil the air is preheated to 400°C, which is used as a reference point in both studied cases. The same temperature is used as the upper limit for the fuel gas (supplementary fuel denoted “NG FUEL” and “OFF-GAS”) which is fed to the reformer as fuel. Excess heat which cannot be used for heating of cold streams (or vented to the atmosphere as in the case with the flue gas) are cooled down in a cooling tower with large cooling water streams. Figure 1 shows the SMR plant including all streams included in the Aspen Plus simulation, stream data can be found in the Appendix.

the presence of oxygen in the fluidization gas which is not present in CLC. In this OCAC process the gaseous fuel can be converted through either a gas-gas reaction with oxygen available in the air or through a gas-solid reaction with the oxygen carrier. The reduced oxygen carrier is then reoxidized through a reaction with gaseous oxygen. The net heat release from the oxidation and the reduction of the oxygen carrier corresponds to the heat of combustion of the gaseous fuel. The process flowsheet for this process (hereafter named case OM since it is based on OCAC and using natural gas as supplementary fuel with methane as main component) can be seen in Figure 2.

Figure 2: Schematic illustration of case OM where the reformer is integrated with a FBHE unit with OCAC. The scheme includes thermal integration with the steam cycle.

3 Model description

3.1 Aspen Plus model design

In this section the main assumptions for the simulated hydrogen production plants are presented where some of the most important assumptions are found in Table 1. The model included the steam reforming process, the thermal integration and the compression of the H_2 product. Peng-Robinson was the base property method for the thermodynamic calculations in Aspen Plus. Solid material such as the oxygen carrier is not included in the Aspen simulation model. The heating value of the natural gas is evaluated using the 'HEATVALS' property set in Aspen Plus. The natural gas stream enters the system at 40 bar, 15°C. The desulfurization is not modelled in Aspen Plus. Sulfur is not included in the NG stream, but in order to account for the need to recycle hydrogen from the product gas to produce H_2S , the model includes a stream of recycled H_2 corresponding to 2 mole-% H_2 in the NG feed stream [45, 56]. The pre-reformer and WGS reactors are modelled as adiabatic reactors whereas the reformer is supplied with heat from the furnace to meet the set outlet temperature (no heat losses are assumed). The pre-reformer, reformer, furnace and WGS reactors are all modelled as RGIBBS equilibrium reactors. In the WGS reactor the hydrocarbons were considered as inert.

It was assumed that there was no pressure drop in the PSA for the separated H_2 product gas, whereas the pressure on the off-gas stream was assumed to be high enough to be supplied to the reformer furnace without compression work. Atmospheric pressure is assumed in the furnace. The S/C ratio is set to 2.7 with a 'Design Spec' in Aspen Plus relating the molar flow of steam to the molar flow of carbon from the natural gas feed stream. The minimum temperature difference between the hot and cold streams was set based on previous work by Spallina et al. [33] and Martínez et al. [34] which was close to the guideline provided in the EBTF-report [57]. The possibility of cost targeting for an optimum minimum temperature difference was excluded from this analysis. The assumptions for the steam cycle were based on the work of Martínez et al. [34] and the EBTF-report [57]. The chosen operating steam pressure is higher than what is used currently in SMR plants today, but in order to compare the different processes in a future scenario it was assumed that more advanced steam cycles with higher efficiencies are used.

It is assumed that the pressure drop reactors is 1% of the inlet pressure over reactors and 2% over all heat exchangers including HRSG and process preheating [33] which is comparable to the assumptions recommended in the EBTF-report [57]. The pressure drop on the water/steam side in the HRSG network was assumed to be 20% over the economisers, 5% over the evaporators and 8% over the superheaters based on the inlet pressure. The system was designed to deliver steam at 92 bar, 485°C resulting in the estimated pressure delivered by the feedwater pump of 132 bar. For the heat balance calculations, the evaporation is assumed to take place at 92 bar. The pump work is small which motivates a simple assumption for the pressure drop on the water/steam side. Apart from the pressure drop over the fluidized bed, no pressure drops are assumed on the fuel and oxidant side.

Table 1: List of assumptions in the simulation model in Aspen Plus of the plants.

Feed streams to SMR plant	
Feed	Natural gas
Supplementary fuel	Natural gas
Natural gas composition [31]	89% CH ₄ , 7% C ₂ H ₆ , 1% C ₃ H ₈ , 0.11% C ₄ H ₁₀ , 2% CO ₂ , 0.89% N ₂ (70 bar and 15 °C)
Lower heating value (LHV) NG MJ/kg	46.49
Air composition (vol-%)	78.08% N ₂ , 20.95% O ₂ , 0.93% Ar, 0.04% CO ₂
Process conditions	
Pre-reforming inlet temperature (°C)	500
Reforming inlet temperature (°C)	620
Reforming outlet temperature, T _{reform,out} (°C)	900
Reforming pressure (bar)	32.3
Steam-to-carbon ratio	2.7
Pressure drops over reactors (% of inlet pressure) [10]	1
H ₂ in inlet stream to desulfurization (mole-%) [8, 9]	2
HT WGS inlet temperature (°C)	330
PSA	
PSA H ₂ separation efficiency (%)	90
Purity in H ₂ stream (%)	100
H ₂ pressure drop over PSA	0
Heat exchangers	
dT _{min} gas-gas/gas-liquid (°C)	20/10
Gas phase pressure drops, % of inlet pressure [10]	2
Heat loss (% of heat transferred) [33]	0.7
Minimum flue gas stack temperature (°C)	150
Air and fuel gas preheat temperature (°C)	400
H₂ compression unit	
Number of stages [34]	1
Isentropic efficiency [34]	82
Mechanical efficiency [34]	99
Final H ₂ conditions (T[°C]/p[bar])	30/70
Steam cycle	
HP steam condition T(°C)/p(bar)	485/92
IP/ LP (steam export) pressure (bar)	34/6
Pressure drops on water/steam side in economizer/evaporator/superheater (% of inlet pressure)	20/8/5
HP isentropic efficiency (%) [31]	92%
IP isentropic efficiency (%) [31]	94%
Mechanical efficiency (%)	98.5%
Pump	
Pump efficiency (%) [31]	70
Driver efficiency (%)	100

The main differences in the models of the reference plant (case A) and the alternative plant configuration (case OM) are listed in Table 2. The outlet temperature of the GFF/FBHE are determined based on an estimation of the heat transfer coefficient in the GFF and FBHE where

equal tube areas are used for the two cases (see section 3.3 for more information). The choice of the assumed air-to-fuel ratio (AFR) is further explained in Stenberg et al. [32]. Compression work to introduce the air is neglected in the GFF but it is included in the FBHE where it is based on the estimated pressure drop over the fluidized bed (see section A.2 in the Appendix for this analysis).

Table 2: List of key differences in the Aspen Plus models of the two plants.

Item	Plant configuration	
	Conv. SMR plant (case A)	NG-OCAC-SMR (case OM)
Process conditions		
Outlet temperature of GFF/FBHE (°C)	1201	935
AFR in furnace	1.09 [58]	1.04 [58]
Air blowers		
Isentropic efficiency (%) [57]	-	85
Mechanical efficiency (%) [57]	-	95

3.2 Reactor design

The reformer tubes are assumed to have a vertical tube orientation in both plants which is the traditional orientation of steam reformer tubes in the industry today. Horizontal tube orientation has been suggested by Rydén et al. [16] and Ortiz et al. [48] but this would increase the risk of gas bypassing within the reformer tubes with solid material accumulating at the bottom of the tubes. A simplified design of the FBHE unit is shown in Figure 3. The hanging tubes would be connected to a pigtail/hairpin connection below the distributor plate into a collection manifold from which the gas then moves up through another reformer tube to make the second pass through the bed.

Figure 3: Illustrative design of a fluidized bed heat exchanger with two passes including connections to the collection manifold.

The overall configurational model of the plants was developed in Aspen Plus, and based on established components. The reformer furnace design calculations are carried out in Excel. Based on assumed reformer tube dimensions (see Table 3), an assumed tube pitch of $2.4D_{to}$ [59] for the gas-fired furnace (GFF) and assuming that distance between the first row of tubes and the tube wall is also equal to $2.4D_{to}$, it is possible to define basic dimensions of the gas fired

reformer furnace. A relation of 2:1 is assumed between the width and the depth for both GFF and FBHE and the reformer tubes are distributed evenly over the available cross section in the reactor. The height of the gas-fired reformer furnace is equal to the length of the vertically placed reformer tube.

The dimensions of the FBHE is on the other hand based on the volumetric flowrate of gas in the furnace estimated in the Aspen Plus model and the limit to maintain a superficial gas velocity of 0.5 m/s which is close to what is used in FBHE units today [14, 60]. It should be pointed out that, the velocities need to be low in order to avoid erosion issues. Since the target is to maximize the heat transfer to the reformer tubes in the FBHE, the reactor is provided with significant refractory lining, an outside insulation and a steel vessel which surrounds the insulation. It is assumed that the required total height of the FBHE is 120% of the bed height to avoid potential loss of particles from the bed.

Table 3: Design assumptions for the reformer tubes [16].

Reformer tube inside diameter, d_{ti}	0.1	m
Reformer tube outside diameter, d_{to}	0.12	m
Reformer tube length, L_t	11.4	m
Equivalent diameter of catalyst pellet, d_{cat} [59]	0.0174	m

Based on the constraints in gas velocity in the FBHE a suitable tube pitch should be decided. It is difficult to define a lower limit for the tube pitch where the lower limit should be connected to a significant reduction of the heat transfer to the tubes. In this work the lower limit was set to a tube pitch corresponding to two reformer tube diameters. This requires therefore at least four passes through the bed where the gas then passes through the bed four times each time with an immersed tube length of $L_t/4$. This corresponds to a tube pitch of approximately $2.1D_{to}$ which was considered as a suitable tube bundle design. In this design the reformer tubes take up approximately 4% of the total bed volume which motivates that the influence of the reformer tubes are neglected in the estimation of the bed voidage. More tube passes would allow for a lower pressure drop with a lower bed height and a more compact fluidized bed heat exchanger, but it does at the same time decrease the estimated bed-to-tube heat transfer coefficient (see section A.2). The pressure drop over the fluidized bed is calculated based on the estimated bed voidage, assumed properties for the bed material (see section 3.3.2 and A.2) and the properties of the flue gas in the unit. The pressure drop over the distributor plate is neglected.

Despite the four-pass configuration, the bed height is still high compared to what is frequently used in BFB. One of the main concerns in this regard could be the gas-solid contact. It should however be noted that the relation between the cross sectional area and the height of the bed is still large. This could point towards a situation with less slugging and vertical internals such as the suggested vertical reforming tubes in this case has been observed to reduce the bubble size significantly [61]. Additional internals [13] (in form of small horizontal tubes) or packing material [62] could also be added to reduce the bubble size. A more detailed design is outside the scope of this paper. The required number of reformer tubes was defined based on having the same thermal input per cross sectional tube area as Rydén et al. [16] which resulted in an estimated residence time of gas of approximately 7 s in the reformer tube which is reasonable for this process. The other reactors and separators, e.g. water gas shift and PSA, were scaled according to literature data.

3.3 Steam reformer heat transfer model

Simulation models of the plant are developed in Aspen Plus, stream data is retrieved and used in the estimation of heat transfer coefficients, calculations which are carried out using Excel

spreadsheets. The result from this calculation results in a new target exit temperature of the furnace in the Aspen Plus and an iterative calculation is carried out. The calculation procedure can be observed in Figure 4.

Figure 4: Calculation procedure to estimate the required flue gas temperature in the reformer furnaces.

3.3.1 Gas fired furnace heat transfer model

The conventional SMR production plant utilize a gas fired furnace in which the heat transfer is largely governed by radiation. To compare the two plant configurations, the differences in heat transfer in the furnace should be accounted for. One approach is to determine the furnace exit temperature in the conventional gas fired furnace where the same number of tubes and tube dimensions are used in case A and case OM. Here, the heat transfer coefficient in the gas-fired furnace to the tube, h_o , was estimated based on the correlations of Gupta et al. [59]. Based on the calculated heat transfer coefficients and the furnace duty, the required heat transfer area can be estimated based on a calculation of the overall heat transfer coefficient ($Q=U_oA_{req}\Delta T_{LM}$). The available heat transfer area (which is the surface area of the reformer tubes) should then be at least as large as the required heat transfer area. The logarithmic mean temperature difference is calculated as the temperature difference between the average furnace temperature (the flue gas temperature from the furnace in the Aspen Plus model) and the temperature at the inlet and outlet of the reformer tube. It should be acknowledged that the temperature profile in the reformer tube is not expected to be linear between 620 and 900°C due to the endothermic steam reforming reaction. Temperature profiles in reformer tubes have been estimated in reformer tube models [59] where a temperature drop close to the inlet can be expected. A temperature drop of approximately 100°C close to the inlet is seen in the results of Gupta et al. [59] but the use of a pre-reformer in this study should result in a lower temperature drop). For simplicity, it is therefore assumed in the reformer tube calculations that the inlet temperature to the reformer is 560°C. This relates only to the heat transfer calculations however. The volumetric flow on the inside of the reformer tube is taken as the average flow of the inlet and outlet gas flow. The furnace duty delivered to the tubes is determined in the Aspen Plus model. The heat transfer coefficient on the inside of the tube, through the tube wall and outside of the tube wall are all estimated in Excel calculations. Based on the available heat transfer surface and a guessed flue gas temperature, iterative calculations are conducted to control at which temperature the available heat transfer surface matches the required heat transfer surface area.

This required flue gas temperature was estimated to 1201°C (see Table 2) in the gas-fired furnace which is a reasonable flue gas temperature. In the reformer furnace the outlet gas compositions was used to determine the thermodynamic properties of the gas in the furnace. The required refractory area in the GFF is calculated using the refractory area used by Gupta et

al. [59] assuming the refractory area (internal insulation) to be directly proportional to the number of reformer tubes.

3.3.2 FBHE heat transfer model

One of the most important properties of the FBHE is the excellent heat transfer from the bed to the immersed tubes. Despite the fact that much work has been done developing correlations to describe the behaviour of the heat transfer in fluidized bed, it is still difficult to make valid predictive models for the bed to tube surface heat transfer [63]. A recent study was conducted to investigate the bed-to-tube heat transfer coefficient at high temperature for a particle size range similar to what is targeted in this work[64]. The results were compared with some of the most well-known bed-to-tube heat transfer correlations where significant differences were observed between the correlations. The estimated heat transfer coefficients were in the range 750-1600 W/(m²K) at bed temperatures ranging from 400-950°C and mean particles sizes of 123-327 µm using air as fluidizing gas. All the correlations included in that study were considered in this work to attempt an estimation of the convective heat transfer coefficient from bed to tube in case OM with the results in the range 497-840 W/(m²K) for all but one correlation which showed significantly higher values. To make a conservative estimation for the heat transfer coefficient, the correlation which showed the lowest estimated bed-to-tube heat transfer coefficient was included in this heat transfer calculations for the FBHE. This correlation was the expression by Andeen & Glicksman [65], see Eq.(22) in the Appendix. The correlation was the result of a modification of the correlation by Vreedenberg for a horizontal tube in a fluidized bed [66]. In these calculations it should be mentioned that the used oxygen carrier in the FBHE was assumed to have a mean particle size of 180 µm and a bulk density of 1600 kg/m³. Additional data for the characteristics of the particles is found in section A.2 in the Appendix. Heat transfer to the tube length which is not immersed in the bed is neglected in this analysis. Eq.(22) was used to find the bed-to-tube heat transfer coefficient for a single horizontal tube. The estimated value is then corrected using correction factors to account for the vertical tube orientation and the effect of the tube bundle. The estimation of the heat transfer coefficient for the FBHE is presented in section A.2 in the Appendix. The procedure for the calculation of the required and the available heat transfer area is repeated for the case with the FBHE as for the GFF to find the targeted flue gas temperature (see Figure 4).

3.4 Thermodynamic assessment

There are numerous possible performance indicators for the process plants since there are multiple outputs from the system. The chemical energy stored in the NG is assumed to be converted into hydrogen, electricity and heat. The possible use of the electricity/heat can be related to the energy required to produce this electricity/heat in a state-of-the-art industrial plant.

Figure 5: Simplified diagram with the streams of mass and energy within the system boundary.

At the same time gases are released to the atmosphere, including CO₂. The approach presented by Martínez et al. [34] is used to compare the performance of the processes. The used parameters are summarized in Table 4.

Table 4: Parameters used in thermodynamic evaluation of the two plants.

Hydrogen production efficiency (%)	$\eta_{H_2} = \frac{\dot{m}_{H_2,prod} \cdot LHV_{H_2}}{\dot{m}_{NG,tot} \cdot LHV_{NG}}$	(3)
Equivalent flow of CH ₄ (mol/s)	$\dot{M}_{CH_4,eq} = \dot{M}_{CH_4} + \frac{7}{4}\dot{M}_{C_2H_6} + \frac{5}{2}\dot{M}_{C_3H_8} + \frac{13}{4}\dot{M}_{C_4H_{10}}$	(4)
Equivalent flow of NG (kg/s)	$\dot{m}_{NG,eq} = \dot{m}_{NG,tot} - \frac{Q_{th}}{\eta_{th,ref} \cdot LHV_{NG}} - \frac{W_{el}}{\eta_{el,ref} \cdot LHV_{NG}}$	(5)
Efficiencies reference plants	$\eta_{th,ref} = 0.9 \text{ [34]} \quad \eta_{el,ref} = 0.583 \text{ [34]}$	
Heat delivered from steam export (W)	$Q_{th} = \dot{m}_{steam,exp} \cdot (h_{sat,vap,6bar} - h_{sat,liq,6bar})$	(6)
Hydrogen yield (%)	$Y_{H_2} = \frac{\dot{M}_{H_2}}{\dot{M}_{CH_4,eq}}$	(7)
Equivalent hydrogen production efficiency (%)	$\eta_{H_2,eq} = \frac{\dot{m}_{H_2} \cdot LHV_{H_2}}{\dot{m}_{NG,eq} \cdot LHV_{NG}}$	(8)
Equivalent hydrogen production efficiency (based on that all LP steam is converted to electricity in a LP steam turbine) (%)	$\eta'_{eq,H_2} = \frac{\dot{m}_{H_2} \cdot LHV_{H_2}}{\dot{m}_{NG,tot} \cdot LHV_{NG} - \frac{W'_{el}}{\eta_{el,ref}}}$	(9)
Conversion factor LP steam turbine	Electricity production LP steam turbine = 0.1 Q_{th}	
Specific CO ₂ emissions from reference plants	$E_{th,ref} = 63.3 \frac{g_{CO_2}}{MJ_{th}} \text{ [34]} \quad E_{el,ref} = 97.7 \frac{g_{CO_2}}{MJ_{el}} \text{ [34]}$	
CO ₂ specific emissions ($g_{CO_2}/MJ_{H_2 \text{ produced}}$)	$E_{CO_2} = \frac{\dot{m}_{CO_2,emitted}}{\dot{m}_{H_2,prod} \cdot LHV_{H_2}}$	(10)
Equivalent CO ₂ specific emissions ($g_{CO_2}/MJ_{H_2 \text{ produced}}$)	$E_{CO_2,eq} = \frac{\dot{m}_{CO_2,emitted} - Q_{th} \cdot E_{th,ref} - W_{el} \cdot E_{el,ref}}{\dot{m}_{H_2,prod} \cdot LHV_{H_2}}$	(11)
Equivalent CO ₂ specific emissions ($g_{CO_2}/MJ_{H_2 \text{ produced}}$)	$E'_{CO_2,eq} = \frac{\dot{m}_{CO_2,emitted} - W'_{el} \cdot E_{el,ref}}{\dot{m}_{H_2,product} \cdot LHV_{H_2}}$	(12)

W_{el} represents the net electricity production of the plant and W'_{el} is the sum of W_{el} and the electricity that could be produced by expanding the available LP steam in a LP steam turbine.

3.5 Economic assessment

The economic analysis is carried out to compare the levelized cost of hydrogen (LCOH) for the two cases according to Eq.(13). The levelized cost calculation includes both capital and operational costs. This paper focuses on the cost to produce hydrogen, no estimation of the profits from selling the hydrogen is made.

$$LCOH \left(\frac{\text{€}}{m^3_{n,H_2}} \right) = \frac{(TPC \cdot CCF) + C_{O\&M,fix} + (C_{O\&M,var} \cdot h_{eq})}{\dot{V}_{H_2} \cdot h_{eq}} \quad (13)$$

The TPC is the total plant cost which includes equipment costs, installation costs, engineering, procurement and construction costs as well as owner's costs and contingencies. Cost escalation and interest during construction are not included. TPC is an overnight cost expressed in base-year (2015) euro. $\dot{V}_{H_2}$ represents the volumetric flow per hour of hydrogen produced in the plant and h_{eq} represents the yearly operating hours of the plant. CCF is the Capital Charge Factor represents distribution of the total plant cost per year over the life time of the plant. CCF can be estimated by aggregating financial assumptions such as loan interest, debt interest, financial

structure, tax structures, etc. to allow for a simple comparison between conventional plants and newly suggested plants. The CCF is for this plant estimated to 14.4 (%/year) based on the method provided by NETL [67] and by using additional data from [68] and [57]. The calculated value is considered to be reasonable based on similar values documented by NETL [67] and Spallina et al. [33] where similar plant constructions were evaluated. No differentiation is done between the two cases in regard to the CCF used in the calculations.

Other important assumptions relating to the economic assessment are provided in Table 5. Assumed prices for electricity and natural gas is based on average prices for industrial consumers in Sweden during the second half of 2015 excluding taxes. The industrial consumers included in these figures had a yearly electricity consumption between 500-2000 MWh and the gas consumption was between 10000-100000 GJ. The average prices across EU-28 are included for comparison.

Table 5: Basic assumptions for economic calculations.

Base year	2015	
CEPCI-index (2015)	556.8	
Dollar-to-euro conversion rate	0.901	US\$/€
LHV NG	46.494	MJ/kg
Plant size	100000	$m_n^3 H_2/h$
Cost NG (Sweden) [69]	11.67	€/GJ
Availability of operating plant (h_{eq}) [11]	7884	hours/year
Plant life time [57]	25	years
Capacity during operation[11]	100	%
Electricity price [69]	59	€/MWh

The economic value of LP steam has been neglected in this analysis since there is generally little demand for this low-value heat source at a refinery (see later discussion in section 4.1). The method to estimate the total cost of the plant is to a large extent based on the presented methodology presented by Manzolini et al. [70] who uses a Bottom-Up Approach (BUA) but for a combined cycle. The target in this method is to obtain an order-of-magnitude cost estimate within +/- 30%.

Table 6: Cost estimation methodology.

Methodology for calculations of the TPC	Cost
Plant component	
Component x	X
Component y	Y
Bare Erected Cost (BEC)	=X+Y
<i>Direct costs as percentage of BEC</i>	
Total installation cost (TIC) [70]	80% of BEC
Total Direct Plant Cost (TDPC)	BEC+TIC
Indirect costs (IC) [70]	14% of TDPC
Engineering procurement and construction (EPC)	TDPC+IC
<i>Contingencies and owner's cost</i>	
Contingency [70]	10% of EPC
Owner's cost [70]	5% of EPC
Total contingencies and owner's cost (C&OC)	15% of EPC
Total plant cost (TPC)	EPC+C&OC

All the above costs are expressed in “base-year” €. The Bare Erected Cost (BEC) includes the cost of process equipment. On-site facilities and infrastructure to support the plant (such as piping/valves, instrumentation etc.), as well as the direct and indirect labour required for construction and/or installation are included in the installation cost. The direct plant cost and the indirect costs together constitute the cost for Engineering procurement and construction. The process contingencies are then added as well as the owner’s cost, which are added to the EPC to constitute the total plant cost. The used contingencies are representative for a mature technology.

The cost of each component is estimated separately. The sizing is based on heat and mass balances. The capacity (S) of each component should be chosen as a parameter that is well connected to the cost. The estimated component costs (C) was derived based on the cost of a reference component C_o with the reference capacity S_o , see Eq.(14).

$$C = m \cdot C_o \left(\frac{S}{mS_o} \right)^n \cdot \frac{CEPCI_{2015}}{CEPCI_{year\ x}} \quad (14)$$

The scale factor is represented as n and m is the number of required units of the reference component. The Chemical Engineering Plant Cost Index (CEPCI) is used to relate costs of reference process equipment (estimated during year x) to the base year in this paper which is 2015 (the first year of capital expenditure) [67].

Table 7: Assumptions for cost estimation of components in both plants used in Eq.(14).

Equipment unit	Scaling parameter	Ref. capacity, S_o	Ref erected cost C_o (M€)	Scale factor, n	Cost year	Units m
Desulfurization	Thermal feed input [MW_{LHV}]	413.8	0.66 [71]	0.67	2011	2
WGS reactor	Thermal feed input [MW_{LHV}]	1246	11.64 [11]	0.67	2007	1
Fluidized bed	Surface area in furnace [m^2]	1748	4.1 [72]	0.6	2014	1
Pre-reformer	Thermal feed input [MW_{LHV}]	1800	17.5 [73]	0.75	2005	1
PSA unit	Molar flow rate [kmol/hr]	17000	34.3 [11]	0.6	2007	1
H ₂ compressor	Compressor power [MW]	26.1	4.15 [74]	0.6	2002	1
Air blower	Compressor power [MW]	1	0.23 [73]	0.67	2005	1
Steam turbine (ST)	ST gross power [MW]	200	33.7 [57]	0.67	2008	1
Pump	Pump power [kW]	197	0.12 [75]	0.6	2008	1

The cost of a fluidized bed reactor is estimated based on cost estimation of a fuel reactor design provided by Bertsch [72] within the EU-financed SUCCESS project [76]. This fuel reactor included reactor shell, insulation and refractory and was part of a large-scale CLC system for power generation (255 MW_e). The equipment cost of the fluidized bed heat exchanger is then estimated by adding up the equipment cost of the fluidized bed and the cost of the reformer tubes. The cost of the distributor plate is neglected in relation to the cost of the rest of the reactor.

The equipment cost of the GFF was estimated based on the cost estimation for a reformer furnace provided in Turton et al. [77] where a cost correlation was presented as a function of the furnace duty (the heat transferred to the reformer tube). Costs of similar magnitude were estimated using the capital cost estimations for reformers provided by Guthrie [78] and Ulrich [79]. The steam turbine cost was estimated by adding up the power production from HP and IP turbine. The cost of the chimney/stack has been omitted in this analysis.

In the flue gas coolers where heat exchange with hot flue gas takes place it is assumed that the overall heat transfer coefficient is equal to 35 W/(m²K) for flue gas-gas systems [33] apart from the case where steam is in the cold stream where 50 W/(m²K) is used [80]. All heat exchangers have been dimensioned neglecting fouling and non-ideal counter current flow. Data from a large-scale ammonia production plant is used to define U-values for syngas coolers and cost data from this plant was also used to estimate the cost of both flue gas coolers and syngas coolers.

In addition to the capital costs, also the Operations & Maintenance costs (O&M) were estimated. These costs include fixed costs which are independent on the availability of the plant such as labour, maintenance and insurance costs. The variable O&M costs are costs related to material which must be added continuously to the process during operation of the plant. The main assumptions are listed in Table 8.

Table 8: List of process O&M costs.

<u>Fixed O&M costs</u>		
Labour costs [71]	1.2	M€
Maintenance cost [70]	2.5	% TPC
Insurance [70]	2	% TPC
<u>Variable O&M costs</u>		
<i>Cooling water</i>		
Cooling water make-up cost [70]	0.35	€/m ³
Pump power for auxiliaries in cooling tower [57]	0.8	% of heat load in condenser to remove excess heat
Evaporative losses [81]	0.8	% of the circulating water flow rate per 5.6°C range
Drift losses [81]	0.001	% of circulating water flow rate
Assumed cooling water range [81]	11	C
Cycles of concentration (measure of water quality) [81]	4	
Blowdown losses = Evaporative losses/(Cycles of concentration-1)		
Loss of cooling water = Evaporative losses + Drift losses + Blowdown losses		
<i>Process water (water mixed with NG)</i>		
Process water cost [70]	2	€/m ³
<i>Oxygen carrier</i>		
Cost manganese ore (raw material) [82]	225	€/ton
Cost for spray drying, calcination, sieving [83]	1000	€/ton
Cost for C28 material	1225	€/ton
Assumed life-time of C28 with gaseous fuel [19]	9000	hours
<i>Pre-reforming and reforming catalyst</i>		
Cost of Catalyst for pre-reforming and reforming [70]	50000	€/m ³
Space velocity in pre-reformer [84]	195	hr ⁻¹
Void fraction [84]	20	%
Catalyst lifetime [70]	5	years
Catalyst attrition rate [70]	10	%/year
<i>WGS reactors</i>		
Catalyst lifetime [11]	315	days
Catalyst cost [11]	499	\$/ft ³
<i>Desulfurization with ZnO</i>		
Space Velocity [84]	40	hour ⁻¹

Cost of Catalyst [85]	355	\$/ft ³
Catalyst attrition rate	10	%/year
Number of units	2	
Void fraction	30	%
Catalyst life time [86]	2.5	years
<i>Internal/external insulation</i>		
Cost of external insulation [82]	1500	€/m ²
Cost of internal insulation (refractory) [82]	900	€/m ²
Life time for insulation	5	years
<i>Reformer tube</i>		
Cost of conventional reformer tube [87]	20000	US\$/reformer tube
Reformer tube life time in GFF	50000	hours
Reformer tube life time in FBHE	100000	hours

The need to replace insulation material has only been considered for the GFF and the FBHE. The amount of catalyst needed in the desulfurizer, pre-reformer and reformer is estimated based on assumed residence time in these units, volumetric flow rates calculated in Aspen Plus and assumed void fractions. Based on the assumption that the amount of WGS catalyst is proportional to the mass flow of gas in WGS step and the quantity of catalyst used in a study of NETL [11] it was possible to estimate the required amount. The cost to replace the catalyst is then based on the time during which it can be used (see attrition rate and life time). In the estimation of the required quantity of catalyst in the desulphurization unit the reactor temperature was assumed to be 370°C. The solid adsorbent material used in the PSA is considered to have a very long lifetime and the make-up cost for those materials are neglected. The process water is calculated as the raw water consumption to the steam reforming and is equal to the water supplied as feedwater subtracting the water recovery from the condensation of the syngas stream. The cost of C28 is estimated by adding up the material cost for manganese ore and manufacturing of the bed material (spray drying, calcination and sieving). The lifetime of the oxygen carrier is based on the loss of fines in the experiments in a 10 kW_{th} unit where the lifetime was estimated to be above 9000 hours [21]. For a desulfurized fuel in a bubbling fluidized bed with relatively low gas velocities this is probably an underestimation. The cost for solid waste disposal of catalysts, oxygen carriers, absorbents, adsorbents, insulation and used equipment is neglected in this analysis.

The cost to replace the high alloy steel reformer tubes were based on the price indication provided by Roberts and Brightling [87] of 20000 US\$/reformer tube (almost 88% of the total equipment cost in case A). This number is surprisingly high and should the rest of the equipment for the GFF constitute a larger share than 12% in practice, it would mean that the difference between case A and OM is smaller than predicted. The reformer tubes are generally designed based on their creep strength for a lifetime of 100 000 hours of operation [87-89]. Due to difficulties to reduce the thermal stress on the reformer tubes the life time in the GFF is however commonly shorter depending on how well the plant is run, here a life time of 50 000 hours is assumed. Based on the reduced risk for hotspots in the FBHE at the reformer tubes and the assumption that the heat flux would be more constant over time in the FBHE it is likely that the thermal stress is smaller in the FBHE. Therefore, it is considered that a downgrading of the tube material to a cheaper solution should be possible or that a longer life time can be assumed. In this model it is assumed that the same tube material is used but with a longer life time of 100 000 hours in the FBHE.

4 Results and discussion

The results of the techno-economic analysis are presented in four different sections. In the first section the thermodynamic performance of the plants is presented and in the second part the reactor design parameters and the results of the heat transfer calculations for both GFF and FBHE are presented. In the third part the economic evaluation is presented. This is followed by a thermodynamic sensitivity analysis where key parameters in the plant configuration are changed and evaluated. Further, this section includes an economic sensitivity analysis where parameters not affecting the thermodynamic performance of the plant are varied.

4.1 Thermodynamic evaluation

The hot and cold composite curves for case A is presented in Figure 6 and for case OM in Figure 7 where both the flue gas section and the syngas section is included.

Figure 6: Hot and cold composite curves for the syngas system and the flue gas system in the conventional SMR process (case A).

Figure 7: Hot and cold composite curves for the syngas system and the flue gas system for case OM.

By comparing the composite curves for case A and case OM, it can be observed that the heat load in the flue gas section is significantly smaller in case OM compared to case A which reduces excess steam production. Even in case OM, there is excess IP steam production in relation to the required feed to the steam reformer. In case A, the feedwater flow to the steam cycle is significantly higher compared to in case OM, which results in that the heat load in the syngas section increases. The total steam production is decreased by 36% from approximately 40.8 kg/s in case A to 26.0 kg/s in case OM. The amount of LP steam to export corresponds to 4.1 kg/s in case OM but as much as 18.8 kg/s in case A.

The thermal efficiency to produce hydrogen and the CO₂ emissions from the two plants is also evaluated where the key results are observed in Table 9.

Table 9: Energy streams and performance parameters of the plant configurations.

		Case A	Case OM
<i>Electricity production/consumption</i>			
Air blowers	MW _{el}		-1.0
H ₂ compressors	MW _{el}	-3.8	-3.8
Steam turbines	MW _{el}	16.9	8.0
Pumps	MW _{el}	-0.8	-0.5
Pumps for heat rejection	MW _{el}	-0.1	-0.2
Net electric power	MW _{el}	12.3	2.6
<i>Heat production/consumption</i>			
Total cooling requirement	MW _{heat}	-12.3	-22.1
Steam export (160°C, 6 bar)	kg/s	18.8	4.1
Furnace duty transferred to reformer tubes	MW	97.9	97.9
<i>Efficiency and emission data</i>			
Hydrogen yield after reformer: Y _{H₂,ref}	%	68.1	68.1
Hydrogen yield after WGS: Y _{H₂,WGS}	%	78.5	78.5
Total NG consumption: $\dot{m}_{NG,tot}$	kg/s	9.10	8.15
H ₂ production efficiency: η_{H_2}	%	70.7	79.0
H ₂ production efficiency: $\eta_{H_2,eq}$	%	83.9	82.1
H ₂ production efficiency: η'_{eq,H_2}	%	75.8	80.2
CO ₂ specific emissions: E_{CO_2}	$g_{CO_2}/MJ_{H_2 \text{ produced}}$	80.7	72.2
Equivalent CO ₂ specific emissions: $E_{CO_2,eq}$	$g_{CO_2}/MJ_{H_2 \text{ produced}}$	68.0	69.5
Equivalent CO ₂ specific emissions: $E'_{CO_2,eq}$	$g_{CO_2}/MJ_{H_2 \text{ produced}}$	75.3	71.1
Total CO ₂ emissions	Mton CO ₂ /year	0.69	0.61

By comparing case A and case OM in Table 9 it can be seen that the electricity output from the turbines is approximately twice as much in case A because of the additional excess heat available in case A. More cooling is required in case OM since less feedwater is used in case OM which otherwise could be used to cool down the syngas stream. The hydrogen production efficiency is 11.6% higher in case OM only considering the output from the plant in form of hydrogen because of lower natural gas consumption in case OM where less supplementary fuel is needed. If both excess heat in form of LP steam and excess electricity production is taken into consideration, case A becomes slightly more efficient. This is mainly due to the significant amount of excess heat in form of LP steam which is available in case A. This can be understood when comparing η'_{eq,H_2} for the different cases where it can be seen that case OM offers a significantly higher production efficiency.

As it was mentioned previously, SMR plants have generally an excess heat production which should be avoided. One example is Preemraff Lysekil, the largest refinery in the Nordic countries which currently has limited use of excess generation of heat/electricity. In this plant, a so called switchable drive configuration is present where pumps/compressors can be driven by electricity or based on mechanical energy generated by the steam turbines [90], while additional electricity production is not required at the plant and there is no known interest to export electricity. Based on the similarity plant size in this work compared to Preemraff Lysekil and the target location of the plant it seems reasonable to consider the use of excess electricity and excess heat in form of LP steam as very limited.

Based on this discussion, η_{H_2} should be regarded as the most important parameter to value in this analysis where case OM is more advantageous in terms of its thermodynamic performance due to the lower natural gas consumptions. When observing the estimated emissions from the plants the conclusions are as expected quite similar. Case OM has lower CO₂ emissions (lower natural gas consumption) while a lower difference exists in case export of both electricity and heat would be possible.

The calculated hydrogen production efficiency for the conventional plant is similar, but some 3 percentage points less compared to previous work on conventional SMR [33, 34]. The difference can be explained by the fact that this work has considered the heat transfer to the reformer tube to estimate the furnace flue gas temperature. The estimated gas temperature was higher than the assumed values in previous works which therefore results in a higher NG consumption in this work and a lower efficiency.

4.2 Reactors design

The reactor design which was carried out for the gas-fired furnace in case A and for the fluidized bed heat exchanger (FBHE) in case OM is presented in Table 10.

Table 10: Heat transfer calculations in the two reformer furnaces.

		SMR (Case A)	NG-OCAC-SMR (case OM)
Bed voidage		-	0.67
<i>Heat transfer coefficient</i>			
Inside, $h_{i,ref}$	W/(m ² K)	1384	1384
Wall, h_w	W/(m ² K)	2872	2872
Outside, h_o			
Convective	W/(m ² K)	0	497
Radiative	W/(m ² K)	138	307
Total outside without correction for tube bundle	W/(m ² K)	-	804
Outside with correction	W/(m ² K)	138	671
Logarithmic mean temperature difference	°C	513	144
Overall heat transfer coefficient (U_o)	W/(m ² K)	118	370
Available (=required) heat transfer area	m ²	1841	1841

It can be observed in Table 10 that the heat transfer coefficient is significantly higher in case OM with a FBHE-unit compared to the conventional plant. This results in turn in a higher overall heat transfer coefficient which enables a lower furnace temperature. In case A it can be observed that the overall heat transfer to the reformer tube is limited by the low heat transfer coefficient on the outside of the tube whereas the heat transfer coefficient on the outside in case OM is approaching the heat transfer coefficient on the inside of the tube.

A sensitivity analysis is also made to see how the heat transfer coefficient affects the relation between the available heat transfer area and the required heat transfer area in the reformer for case OM. This relationship can be seen in Figure 8 where the fluidized bed temperature was assumed to be 935°C (as in case OM) and the relationship between the available heat transfer area and the required heat transfer area was estimated. The heat transfer coefficient estimated for case OM is observed to be on the indicated horizontal line displaying that the available heat transfer area is equal to the required heat transfer area. The corrected heat transfer coefficient

(instead of using the heat transfer coefficient to a single tube) was used in the figure to simplify a comparison with the reference plant.

Figure 8: Variation of corrected heat transfer coefficient on the outside of the reformer tube for case OM at a bed temperature of 935°C.

It can be observed that a significant reduction in the required tube area is obtained when increasing the outside heat transfer coefficient from 138 W/(m²K) which was assumed for the GFF in the conventional SMR plant (noted “Case A” in Figure 8). The effect of having even higher heat transfer coefficients is more limited although not to be neglected. As mentioned previously the results when estimating the convective heat transfer coefficient for case OM with various correlations, the used correlation showed the lowest value out of the studied correlations. Significant benefits are already observed in case OM, but it is possible that even higher heat transfer coefficients could be observed in this system which would make this system even more interesting. The corrected heat transfer coefficient could also be increased by increasing the tube pitch. Another possible benefit with operating with high heat transfer coefficients from bed to tube as in case OM is that it should be easier to control the surface temperature. This is considered since changes in the heat transfer to the tube is expected to have less impact on the surface temperature compared to case A where the heat transfer is more heavily governed by the heat transfer to the tube wall. The design specifications for the reformer furnaces in case A and OM are presented in Table 11.

Table 11: Design results for the reformers (GFF and FBHE)

		Case A	Case OM
Number of reformer tubes		428	428
<i>GFF/FBHE</i>			
Width W	m	8.7	30.0
Depth D	m	4.5	15.0
Height H	m	11.4	3.4
Bed height	m	-	2.85
Pressure drop	bar	-	0.25
Bed inventory	ton	-	1943

Specific bed inventory	kg/MW _{th}	-	19854
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It is easily identified that the footprint of the FBHE unit in case OM is significantly larger than the GFF in the conventional plant. The main reason for this is that the gas velocity is limited to 0.5 m/s. This is a possible drawback with the system since it would take up more space at the plant and larger units result in general in higher capital costs. The required height of the suggested FBHE design including several tube passes is lower than for the conventional reformer but it has a significantly larger cross-section. There are several possible solutions to reduce the cross-section of the FBHE unit such as enabling a bubbling fluidized bed at higher gas velocities by changing to larger and/or more dense bed material. It should however be mentioned that using larger particles for example will reduce the bed-to-tube heat transfer. Another alternative is to divide the unit into different sections where the reformer tubes could be placed in a section with a lower superficial gas velocity and in another section with no tubes a higher superficial gas velocity could be used to potentially reduce the required cross-section of the FBHE unit. It should also be pointed out that a higher flue gas temperature could be used in case OM to reduce the required reformer tube surface area.

4.3 Economic assessment

The results of the economic analysis are presented in Table 12 where the cost of each component is presented as well as the fixed and variable costs to add up to the hydrogen production cost for both cases. In the economic evaluation of the plants, the possibility to export excess electricity is neglected and the excess heat in form of LP steam is regarded to have no economic value (see discussion in section 4.1 for further information). An economic sensitivity analysis is however conducted in section 4.5 to investigate the possible impact of putting a value on these energy streams.

Table 12: Cost comparison between the two plants

Hydrogen production process		Case A	Case OM
<i>Bare Erected Cost (M€ (% of total BEC))</i>			
Desulfurization		0.71 (0.8%)	0.71 (0.8%)
HT WGS reactor		5.32 (6.0%)	5.32 (5.9%)
Reformer furnace		8.82 (9.9%)	10.88 (12.1%)
Pre-reformer		6.16 (6.9%)	6.16 (6.8%)
PSA unit		20.72 (23.3%)	20.72 (23.0%)
H ₂ compressor		1.86 (2.1%)	1.86 (2.1%)
Air blower		-	0.28 (0.3%)
Steam turbine		6.23 (7.0%)	4.17 (4.6%)
Pump		0.24 (0.3%)	0.20 (0.2%)
Syngas cooling HX		10.57 (11.9%)	13.05 (14.5%)
Flue gas cooling HX		24.95 (28.0%)	21.78 (24.1%)
Cooling water system		3.44 (3.9%)	5.10 (5.7%)
Bare Erected Cost (BEC)	M€	89.0	90.2
Total Plant Cost (TPC)	M€	210.1	212.9
Total Plant Cost	M€/year	30.3	30.7
<i>O&M fixed costs (M€ (% of total O&M fixed costs))</i>			
Labour costs		1.20 (8.0%)	1.20 (8.7%)
Maintenance cost		5.25 (35.0%)	5.32 (38.7%)
Insurance cost		4.20 (28.0%)	4.26 (30.9%)
WGS catalyst		0.19 (1.2%)	0.19 (1.3%)
Reformer tube replacement		1.22 (8.1%)	0.61 (4.4%)
Pre-reformer and reformer catalyst		1.31 (8.7%)	1.31 (9.5%)
Desulfurization catalyst		0.33 (2.2%)	0.33 (2.4%)
Internal + external insulation		1.33 (8.8%)	0.56 (4.1%)
Total O&M fixed costs	M€/year	15.0	13.8
<i>O&M variable costs (M€ (% of total O&M variable costs))</i>			
Cooling water make-up		0.06 (0.0%)	0.10 (0.1%)
Process water		0.71 (0.5%)	0.71 (0.5%)
Oxygen carrier make-up		-	2.1 (1.6%)
Natural gas		140.2 (99.5%)	125.6 (97.7%)
Total O&M variable cost	M€/year	140.9	128.5
<i>Hydrogen production cost (including all the above costs)</i>			
LCOH	€/Nm _{H₂} ³	0.236	0.219
LCOH	€/kg _{H₂}	2.629	2.441

It can be observed that the total equipment costs described as the total plant cost is similar for the two plants. One reason for this is that the SMR process is the same for both cases. The NG feed is identical in size for these cases although the total NG consumption is different. One of the differences is the cost of the reformer furnace which is higher in case OM. The equipment cost for the reformer was among the most difficult units to determine the cost for based on lack of published data. As noted previously the cost of the reformer tubes dominate the equipment cost of the reformer and the added cost for the reformer furnace is not substantial. Case OM presents however a lower equipment costs for steam turbines and heat exchangers overall. The heat exchangers for flue gas are however more expensive for case A as expected. The heat exchangers for syngas cooling are more expensive in case OM because of lower temperature difference between hot and cold side. The cooling water system is also more

expensive for case OM in relation to A and a small added cost is observed for the air blower. The O&M fixed costs are similar except for some small differences in the cost for tube replacement and insulation. When considering the variable costs, it can be observed that the make-up cost for oxygen carriers is not negligible in case OM.

As mentioned before, the higher performance of case OM indicates a lower fuel requirement and therefore the cost of the fuel is 10% lower (14.6 M€/year). The revenue for electricity export is not considered, however, this would be equal to 5.7 M€/year for case A and 1.3 M€/year for case OM assuming no additional costs to export the produced electricity. The excess electricity could also have been used to produce additional hydrogen through electrolysis of water. A quick estimation using rather generous assumptions for an electrolysis unit showed however no reduction in the LCOH, mainly due to a high investment cost. The overall cost of plant and LCOH is consistent with previous work [33-35] considering the differences in assumed fuel prices and estimated hydrogen production efficiencies. In summary, the estimated LCOH for case OM is more than 7% lower than the benchmark technology indicating an interesting opportunity for large-scale hydrogen production.

4.4 Sensitivity analysis

A sensitivity analysis was conducted for case OM to calculate the variation in terms of hydrogen efficiency and LCOH based on parameters affecting the thermodynamic modelling of the plant in Aspen Plus. The number of reformer tubes was estimated based on maintaining the same residence time of the gas as in case OM unless stated otherwise. The results are presented in Table 13. The effect of the AFR-ratio was chosen in order to evaluate how important this parameter is for the performance of the system and if this should be seen as another key reason (apart from the bed-to-tube heat transfer) to why case OM is a more interesting plant design than case A. It is expected that a low AFR can be utilized in OM, primarily due to the oxygen buffering of the oxygen carrier particles. The $T_{\text{reform,out}}$, S/C ratio and T_{APH} are considered as some of the most important design parameters for the SMR plant in general and evaluation was done to see if the chosen operating points made sense from a thermodynamic efficiency and LCOH. The residence time was varied to evaluate the effect of using an alternate FBHE design. *Table 13: Results from the sensitivity analysis where the change in % in relation to case OM was estimated for the indicated parameters.*

Air to fuel ratio (AFR=1.04)	η_{H_2} LCOH	AFR = 1.0	AFR = 1.2
		+0.37%	-1.56%
Outlet temperature of the reformer tube ($T_{\text{reform,out}}=900^\circ\text{C}$)	η_{H_2} LCOH	$T_{\text{reform,out}} = 875^\circ\text{C}$	$T_{\text{reform,out}} = 950^\circ\text{C}$
		+0.23%	-2.44%
S/C ratio (2.7)	η_{H_2} LCOH	S/C ratio = 2.35	S/C ratio = 3.5
		-0.40%	-0.15%
Air preheat temperature ($T_{\text{APH}}=400^\circ\text{C}$)	η_{H_2} LCOH	+0.00%	+0.28%
		No air preheating ($T_{\text{APH}} = 36^\circ\text{C}^1$)	$T_{\text{APH}} = 550^\circ\text{C}$
Residence time in reformer tube (ca 7 s)	η_{H_2} LCOH	-7.06%	+0.36%
		+3.49%	+0.67%
	η_{H_2} LCOH	$N_t = N_{t, \text{case OM}/2}$	$L_t = L_{t, \text{case OM}/2}$
		-2.02%	-2.97%
		+0.27%	+0.14%

A lower AFR results as expected in a more efficient hydrogen production and a lower leveled hydrogen production costs. Although this is an added benefit for case OM compared to case A,

¹ Approximate temperature after air blower (assumes no preheating after compression).

it is clear that the change in AFR between case A and OM is not the main reason for the efficiency improvement in case OM. If a higher AFR than the assumed value is needed for case OM, the system should still provide hydrogen production at a lower cost compared to the reference case.

The outlet temperature from the reformer was the second parameter to be studied. A higher outlet temperature results in additional demand for heat to the steam reforming in form of supplementary fuel but at the same time a higher hydrogen yield is obtained in the reformer. Operating with an outlet temperature of 950°C results however in a case where the gain is smaller than the cost in terms of additional supplementary fuel consumption. This case also has a higher equipment cost compared to the base case due to additional requirement of heat exchange surface. At an outlet temperature of around 875°C the thermal input in the off-gas stream is enough to supply all the required heat to the steam reforming reaction and the hydrogen production efficiency is slightly higher. Even though no NG is required as supplementary fuel the production cost is almost identical at the lower temperature in the reformer. The reason for this is mainly a lower hydrogen yield over the reformer resulting in a higher supply of natural gas fed through the reformer tubes which increases the required number of tubes for example. Based on this analysis it therefore makes sense to operate the alternative process at the suggested outlet temperature of 900°C.

The third parameter to be varied was the steam-to-carbon ratio. When the S/C ratio was lowered to 2.35 no supplementary fuel addition is needed and this was therefore set as the lowest studied S/C ratio. The hydrogen yield is higher at higher S/C ratio, but more NG is at the same time required to the furnace due to a higher heat duty in the furnace. It is interesting to see that the hydrogen production efficiency is lower at both 2.35 and 3.5 compared to the base case. At a S/C ratio of 3.5 the benefit from having a higher hydrogen yield in the reformer is smaller than the cost in terms of additional supplementary fuel consumption although the efficiency is only slightly lower. When the ratio is instead 2.35 the benefit from using less supplementary fuel is instead smaller than the cost in terms of reduced hydrogen yield. The equipment cost is lower at a low steam carbon ratio (2.35) which results in a lower cost than the case with high ratio (3.5) mainly based on that less steam needs to be cooled down and heated up resulting in less HX surface area. The LCOH at a S/C ratio of 2.35 was similar to the cost for the base case so a lower S/C ratio could therefore be interesting but operation at a lower S/C ratio also increases the risk of potential issues related to coking.

The fourth parameter to be examined was the amount of air preheating. In the case with air preheating to 550°C the fuel gas was heated as much as possible to maintain the same heat exchanger network and a minimum temperature of the flue gas of 150°C. There is a clear trade-off in this process between having a more thermodynamically efficient process and higher heat exchanger costs or vice versa. At higher temperature, the hydrogen production efficiency increases, reducing the fuel costs, while the heat exchangers costs increase significantly. The level of air preheating which was used in the base case seems suitable.

The fifth parameter to be examined was the residence time where the residence time of the gas is reduced to around 50% in the reformer by using two different measures. In the process where the 50% of the original number of reformer tubes are used the required bed temperature in this case is 998°C which results in an increased demand for NG as supplementary fuel. The cost of the FBHE is however reduced to 7.1 from 10.9 M€ as a result of using less reformer tubes. The hydrogen production efficiency decreases by 2.0% but the LCOH is increased only by 0.3% which indicates that this could be an interesting design alternative, especially if the fuel cost is

lower. In the process where the reformer tube length is reduced by 50% the required flue gas temperature increases to 1026°C. The cost of the reformer tube was assumed to be 50% of the cost of the original tube. The bed inventory and the pressure drop are both reduced but these benefits are small in relation to the price paid in higher NG consumption as a result of lower hydrogen production efficiency. The LCOH is similar to the cost for the base case (only 0.1% higher) mainly as a result of the reduced cost for the FBHE. This shows that alternative FBHE designs could be possible and still enabling a significantly lower production cost compared to the conventional process. In general, it seems that measures to reduce NG consumption are more efficient than targeting lower equipment cost by using a more compact FBHE.

4.5 Economic sensitivity analysis

For the economic sensitivity analysis, another set of parameters were chosen which are indicated in Table 14. In the same table the change in hydrogen production cost is included for case A and OM separately in the chosen ranges.

Table 14: Parameters for the economic sensitivity analysis where studied ranges are indicated from “low” to “high” values representing the lowest and the highest value within the studied range. Both estimated LCOH and price change in % in relation to the respective base case.

Operating parameter	Range	LCOH for Case A			LCOH for Case OM		
		Low	Mid	High	Low	Mid	High
Cost of furnace including reformer tubes (% of estimated cost)	50-200	-1.1% 0.2338	Base	+2.1% 0.2413	-1.4% 0.2163	Base	+2.8% 0.2256
Plant availability (%)	50-100	+18.9% 0.2809	Base	-2.4% 0.2307	20.2% 0.2638	Base	-2.5% 0.2139
Capital Charge Factor (CCF) (%/year)	5-20	-10.6% 0.2112	Base	+6.3% 0.2511	-11.6% 0.1940	Base	+6.8% 0.2344
Reformer tube lifetime (hours)	25000-100000	+0.7% 0.2379	Base	-0.3% 0.2355	+1.1% 0.2217	-	Base
Price for excess electricity (€/MWh)	0-120	Base	-	-6.2% 0.2215	Base	-	-1.4% 0.2163
Price of LP steam (€/ton)	0-40	Base	-	-11.6% 0.2088	Base	-	-2.7% 0.2135
Natural gas price (€/GJ)	5-20	-43.0% 0.1347	Base	+53.7% 0.3632	-41.5% 0.1284	Base	+51.8% 0.3333
Oxygen carrier price (% of assumed price)	0-200	-	-	-	-1.2% 0.2168	Base	+1.2% 0.2221
Cost to emit CO ₂ (€/ton)	0-100	Base	-	+36.8% 0.3233	Base	-	+33.5% 0.3102

It can be observed that the cost of the furnace as well as the tube lifetime has small influence on the levelized production cost in relation to other parameters. It should however be mentioned that this is the case assuming no change in availability of the plant. When the availability of the plant is varied it can be seen it has a significant impact on LCOH, in particular when the plant availability is reduced to 50%. Therefore, to compete with the conventional SMR (whose availability is normally >90%.) a similar availability is required. The availability for case OM must be at least 69% to have a lower LCOH than in case A. Since almost all components are identical in the two plants, the difference in availability should depend on the operability of the reformer furnace which is difficult to assess at this stage. Changes in the cost of the reformer tubes (here illustrated as a change in the tube lifetime) does have an impact on the production cost. It is difficult to say in advance what reformer tube lifetime can be expected in the two cases, but these results show that measures to increase the reformer tube lifetime is still important to reduce the LCOH. However, the influence of the reformer tube lifetime on the plant availability is probably even more important.

In case the electricity and steam-to-export are considered as valuable products, the LCOH of case A is closer (or higher) than for case OM. It should however be mentioned that the additional costs of infrastructure for export of electricity such as generators and steam network for possible export of LP steam are not included in this analysis. In a recent study the cost of the accessory electricity plant was estimated for a conventional SMR plant for hydrogen production where this plant constituted close to 15% of the total equipment cost [91]. This indicates that the investment cost for the additional equipment to export electricity could be significant. In case of low electricity price (as in Sweden), the benefit is significantly reduced. The value of LP steam is considered to be in the lower part of the studied range at a refinery in Sweden where presence of decentralized district heating for example could enable some economic benefits.

As anticipated, the cost of natural gas is the most sensitive parameters for the final LCOH. Sweden has a high natural gas price and therefore a more efficient plant – as in the OM case – will be more convenient. The make-up cost for oxygen carrier is not negligible and possibilities to use cheaper oxygen carriers should be considered if high fuel conversion and mechanical stability can still be maintained. It is for example observed that the specific bed inventory is high compared to many experimental studies in CLC. At such high specific bed inventories and in presence of gaseous oxygen the choice of the oxygen carrier may not be limited by reactivity which also could motivate using a cheaper material based on iron or manganese for example. In presence of a carbon tax of 100 €/ton (as could be expected in the future) the LCOH is expected to increase more than 30% for both cases but due to more CO₂ emissions in case A, the cost increase is larger for this case in the absolute numbers.

5 Conclusions

An integrated fluidized bed heat exchanger (FBHE) for H₂ production in a steam methane reformer using natural gas as feed and supplementary fuel has been assessed and compared with conventional SMR plant using a gas-fired furnace from a techno-economic point of view. The alternative plant configuration has a significantly lower variable cost (which is dominating the levelized hydrogen production cost) resulting from a 11.6% higher hydrogen production efficiency η_{H_2} . This in turn originates from the improved heat transfer in the furnace to the steam reforming tubes. The capital and fixed costs are similar for the two plant configurations resulting in a 7.1% lower LCOH for the alternative process configuration and the excess steam production is reduced by 36%. The total plant cost and the fixed costs of the plants are similar. In case LP steam-to-export and excess electricity production are limited and not valuable the presented process is more convenient from both a thermodynamic, economic and environmental point.

The sensitivity analysis displayed several trade-offs in the plant where reduced fuel consumption often competes with reduced heat transfer area and the importance of each parameter was evaluated. Regardless of how those choices are made, the alternative process configuration with a FBHE unit in the reformer furnace should be a very interesting case for a large-scale hydrogen production. The plant configuration would look very similar to the conventional plant and therefore the existing plant could also be retrofitted by changing the reformer furnace from a gas-fired furnace to a fluidized bed heat exchanger.

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Appendix

A.1 Estimation of heat transfer in GFF

The temperature of the radiating gas was not considered to be the same as the furnace exit temperature here as opposed to the case with a fluidized bed where this is a more reasonable assumption. In the model developed by Rydén et al. [16] to model the heat transfer in the gas fired furnace, the temperature of the radiating gas was determined to be approximately 1377°C based on calibration of the resulting temperature profile over the reformer tube in the work of Gupta et al.[59]. Based on this value and by using the assumptions adopted by Gupta et al. [59], the heat transfer in the furnace could be determined (see [59] for additional details on the calculation). The radiative heat transfer is however dependent on the tube wall temperature which in turn is estimated by considering the temperature in the reformer to be equal to the assumed average temperature in the reformer tube. The heat transfer coefficient inside the tube is therefore first estimated which is used to determine the temperature on the inside of the reformer tube wall. The heat transfer through the tube wall is then used to estimate the temperature on the outside of the reformer tube wall. The heat transfer coefficient from the inside of the reformer tube can be calculated according to Eq.(15) and (16) [92].

$$h_{i,ref} = \frac{k_{g,reform}}{d_{cat}} [2.58 Re_{cat}^{\frac{1}{3}} Pr_{g,reform}^{\frac{1}{3}} + 0.094 Re_{cat}^{0.8} Pr_{g,reform}^{0.4}] \quad (15)$$

$$Re_{cat} = \frac{d_{cat} G_{reform}}{\mu_g} \quad (16)$$

Gas properties are evaluated separately at the assumed inlet temperature of 560°C and the outlet temperature of 900°C and an average of those values is used in the estimation of the heat transfer coefficient inside the reformer tube. The heat flux to the inside of the reformer tube (which is known from the calculated heat transferred to the reformer tubes in the simulation model) can be used to determine the inside wall temperature [92].

$$q_{w,i} = 0.4 h_{i,ref} (T_{w,i} - T_{ref}) \quad (17)$$

The temperature on the outside of the tube wall can be estimated according to Eq.(18) [92] and Eq.(19) [59] since $q_{w,o} = q_{w,i} * (d_i/d_o)$.

$$q_{w,o} = \frac{2k_w(T_{w,o} - T_{w,i})}{d_{t,o} \ln \left(\frac{d_{t,o}}{d_{t,i}} \right)} \quad (18)$$

The thermal conductivity of the tube wall is calculated according to Eq.(19) where T_w is the average of the inside and outside wall temperatures.

$$k_w = 10.738 + 0.0242 T_w \quad (19)$$

The parameter expressed in this article as the wall heat transfer coefficient is determined according to Eq. (20).

$$h_w = \frac{2k_w}{d_{t,o} \ln \left(\frac{d_{t,o}}{d_{t,i}} \right)} \quad (20)$$

The overall heat transfer coefficient is estimated according to Eq.(21).

$$U_o = \frac{1}{h_o} + \frac{d_{t,o} \ln \left(\frac{d_{t,o}}{d_{t,i}} \right)}{2k_w} + \frac{d_{t,o}}{d_{t,i}} \frac{1}{h_{i,ref}} \quad (21)$$

This was then used to estimate the required heat transfer area using $Q=U_o A_{req} \Delta T_{LM}$.

A.2 Estimation of heat transfer in FBHE

In the expression both the convective and radiative heat transfer to the tube surface is considered [15, 63].

$$h_{o,FBHE} = 900(1 - \varepsilon_b) \frac{k_g}{d_t} \left[\frac{u_0 d_{to} \rho_p}{\mu_g} \frac{\mu_g^2}{d_p^3 \rho_p^2 g} \right]^{0.326} Pr^{0.3} + \frac{\sigma(T_b^4 - T_{w,o}^4)}{\left[\frac{1}{e_b} + \frac{1}{e_w} - 1 \right] (T_b - T_{w,o})} \quad (22)$$

The correlation assumes maximum radiative heat transfer meaning that the particles contributing to the radiation are assumed to have the same temperature as the bed. This may not be entirely true for small particles when the particles cool off faster in contact with the tube surface. If the particles that are cooling down are exchanged at a rate which is fast enough, this effect is however small which is assumed in this expression. The bed emissivity (ε_b) is estimated based on the particle emissivity (ε_p) [93].

$$e_b = 0.5(1 + e_p) \quad (23)$$

The bed voidage is an important parameter in the expression to relate the change in the heat transfer with the gas velocity and it was observed by Grewal that his correlation for bed voidage better fit the available data [94] when using Eq.(24).

$$\varepsilon_b = \frac{1}{2.1} \left[0.4 + \left[4 \left(\frac{u_0 \mu_g}{d_p^2 \rho_p \phi_p^2 g} \right)^{0.43} \right]^{0.33} \right] \quad (24)$$

Additional assumptions to complete the estimation of the heat transfer coefficient in the FBHE are observed in Table 15.

Table 15: Design assumptions for fluidized bed heat exchanger and the fluidized bed particles.

Fixed bed voidage, ε_{fixed}	0.4
Particle sphericity, ϕ_p	0.8
Emissivity of the wall, e_w	0.9
Emissivity of the particles, e_p	0.925

The above expression for the heat transfer coefficient does not account for the impact of the tube bundle on the bed voidage and the expression also assumes a horizontal orientation of the tube surface. Basu [15] provides correction factors to account for the use of vertical tubes as well as the distance between the wall and the centerline of the tubes (ξ_1) and the tube pitches (ξ_2), see Eq.(25).

$$h_{o,FBHE,corrected} = \xi_1 \xi_2 h_{o,FBHE} \quad (25)$$

A.3 Stream data from Aspen Plus simulation

Table 16: Stream data for case A

[illegible]

Table 17: Stream data for case OM

[illegible]

List of figure captions

Figure 1: Schematic illustration of the conventional SMR process having a gas-fired furnace (GFF) fuelled with natural gas (NG) including the thermal integration with the steam cycle...	8
Figure 2: Schematic illustration of case OM where the reformer is integrated with a FBHE unit with OCAC. The scheme includes thermal integration with the steam cycle.....	9
Figure 3: Illustrative design of a fluidized bed heat exchanger with two passes including connections to the collection manifold.....	12
Figure 4: Calculation procedure to estimate the required flue gas temperature in the reformer furnaces.	14
<i>Figure 5: Simplified diagram with the streams of mass and energy within the system boundary.</i>	15
Figure 6: Hot and cold composite curves for the syngas system and the flue gas system in the conventional SMR process (case A).	21
Figure 7: Hot and cold composite curves for the syngas system and the flue gas system for case OM.	22
Figure 8: Variation of corrected heat transfer coefficient on the outside of the reformer tube for case OM at a bed temperature of 935°C.	25

List of table captions

Table 1: List of assumptions in the simulation model in Aspen Plus of the plants.	11
Table 2: List of key differences in the Aspen Plus models of the two plants.	12
Table 3: Design assumptions for the reformer tubes [16].	13
Table 4: Parameters used in thermodynamic evaluation of the two plants.	16
Table 5: Basic assumptions for economic calculations.....	17
Table 6: Cost estimation methodology.....	17
Table 7: Assumptions for cost estimation of components in both plants used in Eq.(14).	18
Table 8: List of process O&M costs.....	19
Table 9: Energy streams and performance parameters of the plant configurations.	23
Table 10: Heat transfer calculations in the two reformer furnaces.	24
Table 11: Design results for the reformers (GFF and FBHE).....	25
Table 12: Cost comparison between the two plants.....	27
Table 13: Results from the sensitivity analysis where the change in % in relation to case OM was estimated for the indicated parameters.....	28
Table 14: Parameters for the economic sensitivity analysis where studied ranges are indicated from “low” to “high” values representing the lowest and the highest value within the studied range. Both estimated LCOH and price change in % in relation to the respective base case. .	30
Table 15: Design assumptions for fluidized bed heat exchanger and the fluidized bed particles.	34
Table 16: Stream data for case A	35
Table 17: Stream data for case OM.....	36

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