



# Chemical Looping Technologies for H<sub>2</sub> Production with CO<sub>2</sub> Capture

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## Chemical Looping Technologies For H<sub>2</sub> Production With CO<sub>2</sub> Capture: Thermodynamic Assessment And Economic Comparison

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### Abstract

This work addresses the techno-economic assessment of two chemical looping technologies for H<sub>2</sub> production from natural gas fully integrated with CO<sub>2</sub> capture. In the first configuration, chemical looping combustion operated with a dual circulating fluidized bed system at atmospheric pressure is used as furnace for the reforming reaction. In the second configuration, a chemical looping reforming system at pressurized conditions is used for the production of the reformed syngas. Both configurations have been designed and compared with reference technologies for H<sub>2</sub> production based on conventional fired tubular reforming with and without CO<sub>2</sub> capture.

The results of the analysis show that both new concepts can achieve higher H<sub>2</sub> reforming efficiency than a conventional plant when integrated with CO<sub>2</sub> capture (+8-10% higher). The improvement in the performance of the plant is accompanied with an efficiency penalty of 4-6% and the cost of CO<sub>2</sub> avoidance varies from 20-85 €/ton<sub>CO2</sub>.

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*Keywords:* chemical looping; hydrogen production, CO<sub>2</sub> capture, energy analysis, steam reforming

### 1. Introduction

This work reports a techno-economic assessment of two chemical looping technologies for H<sub>2</sub> production from natural gas fully integrated with CO<sub>2</sub> capture. H<sub>2</sub> represents an important product for the chemical industry [1], and

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natural gas steam reforming is the established process for H<sub>2</sub> production [2], which uses a multi-tubular fixed bed reactor (fired tubular reformer-FTR) to convert natural gas into syngas where the heat of reaction is provided via an external furnace. Alternatively air or oxygen is added in order to reach auto-thermal operation (ATR) [2–5]. When integrating CO<sub>2</sub> capture, different separation systems may be considered for different emission sources. The first emission source is CO<sub>2</sub> in the syngas, which is typically separated by MDEA scrubbing, prior to purification in the PSA-unit leading a carbon capture ratio (CCR) of 60% [6–9]. In order to capture the CO<sub>2</sub> from the flue gas of the furnace and achieve overall capture efficiencies above 85-90%, it is possible to either use part of the decarbonized hydrogen as fuel in the furnace [9] or include a post-combustion capture unit at the gas stack of the reformer by MEA absorption [10].

Among the several solutions already proposed for CO<sub>2</sub> capture, chemical looping technologies are among the most promising and efficient alternatives, since the CO<sub>2</sub> separation is inherently integrated in the fuel conversion step [11]. In chemical looping systems, a metal oxide (named oxygen carrier, OC) is oxidized with air and reduced by converting a fuel into CO<sub>2</sub>/H<sub>2</sub>O (chemical looping combustion, named CLC) or syngas (chemical looping reforming, named CLR). CLR consists of two reactors operated at high pressure in which the oxygen carrier and catalyst are circulated to transfer the oxygen and heat for the reforming reaction from the air reactor (where the exothermic reaction occurs) to the fuel reactor. Many authors [12,13] have proposed novel approaches to integrate CLC and steam reforming, where the chemical looping reactors act as combustion chamber to provide the heat of reaction to the reforming tubes which are immersed into the fuel reactor which converts the PSA-offgas into CO<sub>2</sub>/H<sub>2</sub>O in the fuel reactor.

In this paper, firstly the plants are described for two different chemical looping configurations, referred to as SMR+CLC and CLR. After defining the key performance indicators, an energy and economic analysis of the two configurations is performed. The results are compared with two benchmark technologies based on steam reforming technology: the first option is related to the conventional technology currently used in the industry (referred to as SMR), while the second technology also include CO<sub>2</sub> capture using chemical absorption and H<sub>2</sub>-rich fuel for the combustion in the furnace to supply the heat of reaction to the reformer (here referred to as SMR+CA).

## Nomenclature

AR	Air Reactor
ATR	Auto-Thermal Reforming
BEC	Bare Erected Cost, M€
CA	Chemical Absorption
CCF	Capital Charge Factor
CCR	Carbon Capture Rate
CLC	Chemical Looping Combustion
CLR	Cost of Hydrogen, €/Nm <sup>3</sup>
E <sub>CO<sub>2</sub></sub>	CO <sub>2</sub> specific emissions, kg <sub>CO<sub>2</sub></sub> /Nm <sup>3</sup> <sub>H<sub>2</sub></sub>
FR	Fuel Reactor
FTR	Fired Tubular Reforming
MEA	Mono ethanolamine
MDEA	methyl diethanolamine
OC	Oxygen carrier
HR	Heat Rate, Gcal/kNm <sup>3</sup> <sub>H<sub>2</sub></sub>
PSA	Pressure Swing Adsorption
SPECCA	Specific Primary Energy Consumption for CO <sub>2</sub> avoided
SMR	Steam Methane Reforming
TEC	Total Equipment Cost, M€
TOC	Total overnight cost, M€
WGS	Water Gas Shift

## 2. Description of the plant

### 2.1. Steam Methane Reforming with Chemical Looping Combustion (CLC+SMR)

In the first configuration, named CLC+SMR (**Figure 1**), the natural gas (A01) is first heated-up and sent to the desulphurization unit where H<sub>2</sub>S is removed using ZnO. The natural gas is then mixed with H<sub>2</sub>O from the steam turbine and heated to 500 °C and fed to the adiabatic pre-reformer (A02). The pre-reforming is required in order to convert the high hydrocarbons into CH<sub>4</sub> and reduce the heat duty of the reforming section. The syngas is then heated to 620 °C and fed to the steam reforming tubes. In the CLC+SMR configuration, the steam reformer (SMR) tubes are located inside the fuel reactor (FR) of a CLC system. The heat of reaction is provided by the solids circulation (A15-A16 loop). The fuel reactor is operated in the bubbling fluidization regime and the temperature inside the reactor is considered uniform (above 50 °C compared to the maximum reforming temperature). The reformed syngas is then converted through a HT-WGS reactor (at 400 °C) to enhance the H<sub>2</sub> yield and the H<sub>2</sub>-rich syngas is then cooled, H<sub>2</sub>O is condensed and the dry gas (A05) is sent to the PSA to recover pure H<sub>2</sub> (99.999%) [13]. The PSA tail gas (A08), rich in CO<sub>2</sub>, H<sub>2</sub> and unconverted CO and CH<sub>4</sub>, is pre-heated and sent to the fuel reactor, where it reacts with an oxygen carrier (OC) to form CO<sub>2</sub> and H<sub>2</sub>O. The exhausts (A09) from the fuel reactor are cooled down supplying the heat for the natural gas pre-heating and producing HP steam for power generation. The CO<sub>2</sub> is finally separated from the H<sub>2</sub>O and sent for compression up to 110 bar (A07). The air (A10) is heated to 400-500 °C and fed to the air reactor (AR) where the oxygen carrier is completely oxidized. The high temperature O<sub>2</sub> depleted air (A11) is then cooled down supplying heat to the syngas to the reformer, producing HP steam for electricity production and finally in a Ljungstrom-type heat exchanger for air pre-heating. The steam cycle is operated using a HP steam generator at 100 bar. The steam turbine (ST) inlet conditions (A13) are 485 °C and 92 bar. During the syngas expansion, part of the steam is extracted and sent to the process in order to reach the required S/C ratio. The remaining steam is finally expanded to 6 bar and used as steam export (A14).

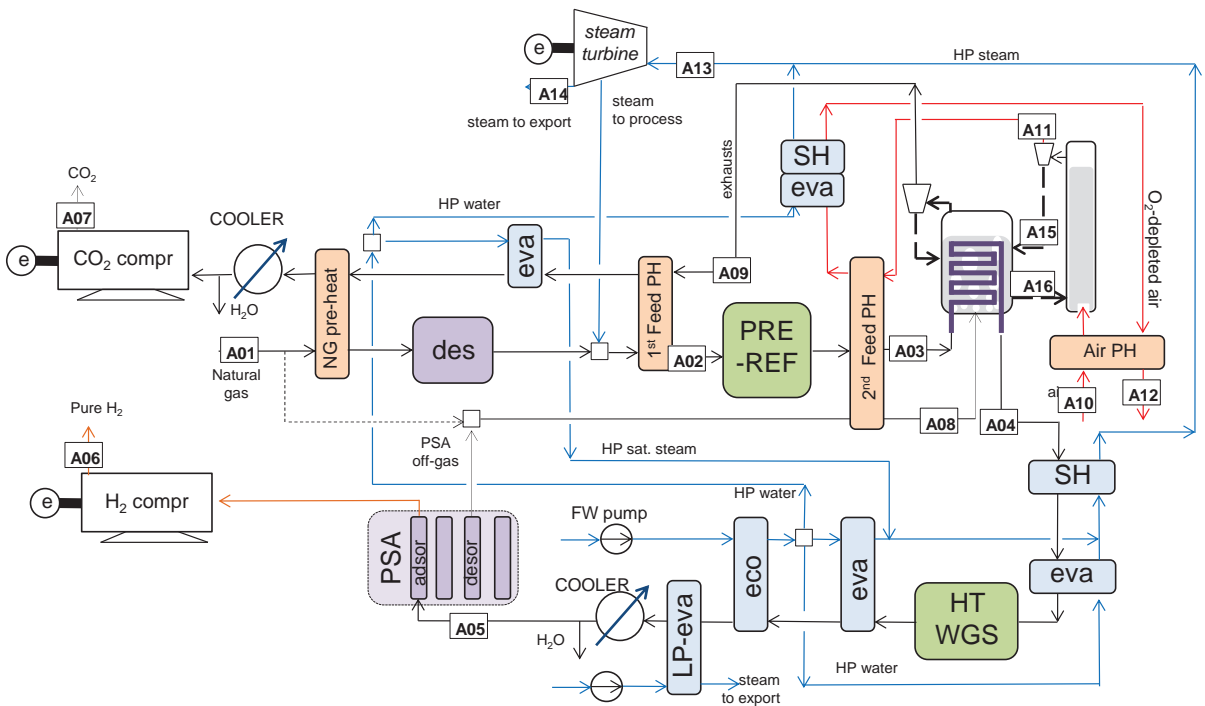


Figure 1: Schematic of Chemical Looping Combustion integrated with Steam Methane Reforming (CLC+SMR)

## 2.2. Chemical Looping Reforming (CLR)

The second configuration (**Figure 2**) is based on chemical looping reforming (CLR). With respect to the conventional Auto Thermal Reforming operated with pure oxygen, this configuration does not require a dedicated air separation unit, because the oxygen is separated using the oxygen carriers. The natural gas is converted in the fuel reactor of a pressurized interconnected fluidized bed system reacting with steam (S/C between 1.5 and 2) and the OC. Compared to the CLC+SMR, this configuration integrates steam reforming and chemical looping in the same unit. The produced syngas (B04) is cooled down and sent to two WGS reactors operated in series at two different temperatures in order to enhance the H<sub>2</sub> yield of the process and increase the CO<sub>2</sub> concentration. The H<sub>2</sub>/CO<sub>2</sub> rich stream (B05) is then sent to an ammine scrubber operated with MDEA where most of the CO<sub>2</sub> is separated and sent to compression and final storage (B07). The H<sub>2</sub>-rich syngas is finally fed to a PSA unit in order to improve the H<sub>2</sub> purity to meet the requirements and compressed up to 110 bar (B06). The PSA-off gas (B08) is combusted in an auxiliary post-combustor to recovery additional thermal power for the reactant pre-heating and to produce additional steam for the steam cycle. The air reactor of the chemical looping unit is fed with compressed air (B09) from a dedicated gas turbine and the HT-N<sub>2</sub> stream from the air reactor (B10) is expanded and used for the heat recovery of the system. This plant is operated with lower S/C ratio than the CLC+SMR because the oxygen carrier also takes part in the reforming reactions, resulting in reduced steam consumption.

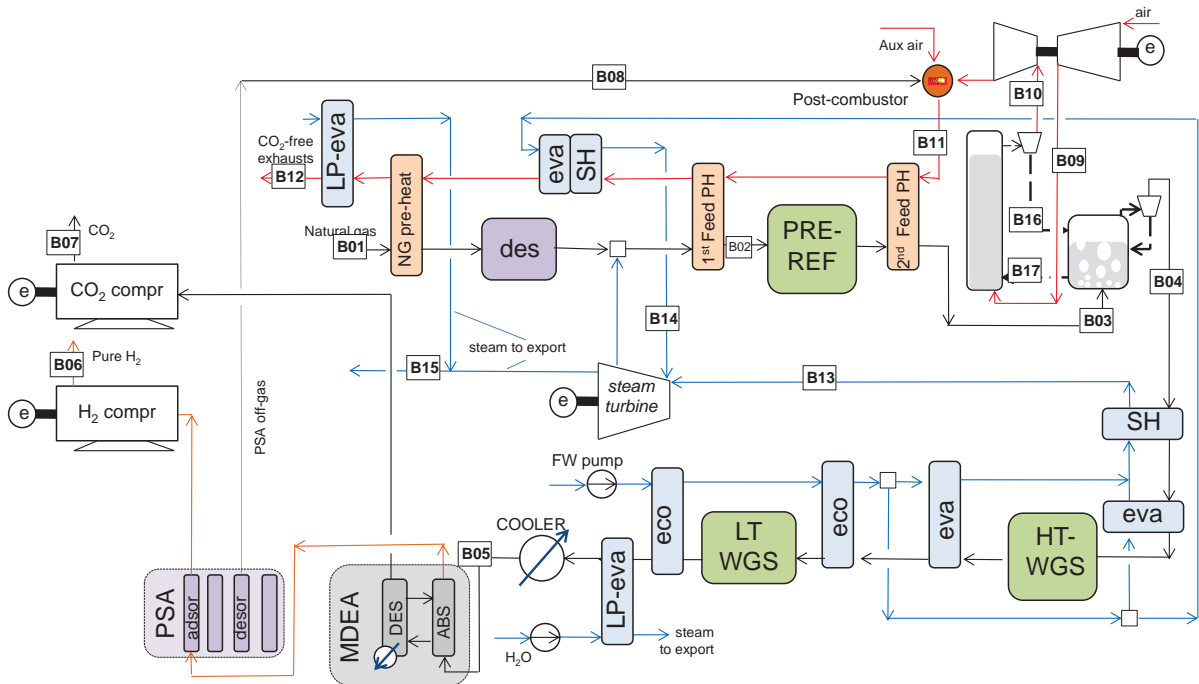


Figure 2: Schematic of Chemical Looping Reforming (CLR)

## 2.3. Main Assumptions and Methodology

The main assumptions used for the simulations have been taken from Spallina et al. [14], Martinez et al. [9], and the EBTF report [15]. The reactor design for the chemical looping reactors has been carried out using a two-phase phenomenological fluidized bed reactor model for the freely bubbling fluidization regime at steady state conditions.

Each plant will convert the chemical energy from the natural gas into H<sub>2</sub>, electricity and heat (as steam export) and will release to the environment part of the CO<sub>2</sub> while the remaining part will be captured and stored. Following Martinez et al. [9] different indices have been chosen to quantify the plant performance.

$$\text{Equivalent natural gas flow rate} \quad \dot{m}_{NG,eq} = \dot{m}_{NG} - \frac{Q_{th}}{\eta_{th,ref} LHV_{NG}} - \frac{W_{el}}{\eta_{el,ref} LHV_{NG}} \quad (1)$$

$$\text{where } \eta_{th,ref} = 0.9; \eta_{el,ref} = 0.583$$

$$\text{Steam export} \quad Q_{th} = \dot{m}_{steam,export} (h_{steam@6bar} - h_{liqsat@6bar}) \quad (2)$$

$$\text{H}_2 \text{ production efficiency} \quad \eta_{H_2} = \frac{\dot{m}_{H_2} LHV_{H_2}}{\dot{m}_{NG} LHV_{NG}} \quad (3)$$

$$\text{equivalent H}_2 \text{ production efficiency} \quad \eta_{H_2,eq} = \frac{\dot{m}_{H_2} LHV_{H_2}}{\dot{m}_{NG,eq} LHV_{NG}} \quad (4)$$

$$\text{CO}_2 \text{ specific emissions (E}_{CO_2}\text{)} \quad E_{CO_2} = \frac{\dot{m}_{CO_2,capt}}{\dot{m}_{NG} LHV_{NG} E_{NG}} \quad (5)$$

$$\text{equivalent CO}_2 \text{ specific emissions (E}_{CO_2,eq}\text{)} \quad E_{CO_2,eq} = \frac{\dot{m}_{CO_2,capt} - Q_{th} E_{th,ref} - W_{el} E_{el,ref}}{\dot{m}_{NG} LHV_{NG} E_{NG}} \quad \text{where} \quad (6)$$

$$E_{th,ref} = 63.3 \left[ \frac{g_{CO_2}}{MJ_{th}} \right]; E_{el,ref} = 97.7 \left[ \frac{g_{CO_2}}{MJ_{el}} \right]$$

$$\text{Energy consumption or Heat Rate} \quad HR_{tot} = \frac{\dot{m}_{NG} LHV_{NG} - Q_{th} - W_{el}}{\dot{N}_{H_2} 22.414} \left[ \frac{Gcal_{NG}}{kNm^3_{H_2}} \right] \quad (7)$$

$$\text{Cost of Hydrogen} \quad COH = \frac{(TOC \cdot CCF) + C_{O\&M,fix} + (C_{O\&M,var} h_{eq})}{\dot{N}_{H_2} 22.4143600 \cdot h_{eq}} \left[ \frac{\text{€}}{Nm^3_{H_2}} \right] \quad (8)$$

### 3. Results

#### 3.1. Analysis of performance

The energy balances of the two novel H<sub>2</sub> production plants is presented in Table 1. Compared to the reference technology (without CO<sub>2</sub> capture), the CLC+SMR plant achieves almost the same reforming efficiency (and therefore H<sub>2</sub> yield). The overall net electricity production is lower because of CO<sub>2</sub> compression, while the other units are slightly different. The use of CLC for the treatment of the PSA offgas is able to capture 100% of the CO<sub>2</sub> produced in the plant, and, based on the production of high steam to export, the CO<sub>2</sub> equivalent emissions are negative. The overall HR is slightly higher (+1.3%) than the HR of the conventional SMR demonstrating the low energy cost for CO<sub>2</sub> capture.

In case of CLR, the H<sub>2</sub> reforming (and H<sub>2</sub> yield) is lower than CLC+SMR (70% vs 73%). The lower H<sub>2</sub> production is partly compensated by the lower electricity demand, but also a lower steam export is obtained. This is reflected in both the HR<sub>tot</sub>, which is 5.7% higher than in the case of SMR (without CO<sub>2</sub> capture), and the CCR (based on equivalent CO<sub>2</sub> emissions), which is 93%.

Both plants show a better performance than SMR integrated with CO<sub>2</sub> capture. In the case of CLC+SMR, the higher H<sub>2</sub> yield is due to the fact that the furnace does not need H<sub>2</sub> as fuel. The higher steam to export (in case of

CLC+SMR) is due to the fact that no steam is required in the reboiler of the amine scrubber. In case of CLR, also the electricity consumption is lower.

A sensitivity analysis has been carried for both CLC+SMR and CLR. In the first case, varying the reforming temperature (800-1000 °C), the H<sub>2</sub> yield decreases, while the equivalent H<sub>2</sub> production increases because of lower energy consumption and higher steam to export. No significant effect has been found when varying the pressure (20-50 bar) due to the fact that at higher CH<sub>4</sub> conversions in the reformer less fuel is available with the PSA off-gas and therefore auxiliary NG is required as fuel for the FR. In case of CLR, an increase in the reforming temperature (from 850 to 950 °C) implies a reduction in the H<sub>2</sub> yield because more fuel is converted to heat the reactants, however the equivalent H<sub>2</sub> efficiency increases because of increased power generation in the gas turbine (the TIT increases because the AR temperature increases). The same trend observed at 32 bar is obtained at lower pressures of 20 bar which is preferred for the gas turbine. In particular, at 32 bar the difference between  $\eta_{H_2}$  and  $\eta_{H_2eq}$  increases.

Table 1: Energy balance and list of performance

		SMR	SMR	CLC+SMR	CLR
CO <sub>2</sub> capture		N/A	CA-MDEA		
Conditions (T/p)		from Spallina[14]		900/32	900/20
inlet fuel	kg/s	2.62	2.62	2.62	2.62
Thermal Input	MW <sub>LHV</sub>	121.94	121.94	121.94	121.94
pure H <sub>2</sub>	kg/s	0.75	0.70	0.75	0.72
	Nm <sup>3</sup> /h	30259	28211	30101	28899
$\eta_{H_2}$	%	74	69	73	70
H <sub>2</sub> yield	mol <sub>H<sub>2</sub></sub> /mol <sub>NG</sub>	2.49	2.32	2.44	2.34
Electricity	MW <sub>e</sub>				
Gas Turbine		-	-	-	1.25
Steam Turbine		3.27	3.53	3.15	3.56
Air fan		0.68	0.85	0.56	-
pumps		0.21	0.27	0.20	0.21
H <sub>2</sub> compressors		2.27	2.13	2.25	2.77
CO <sub>2</sub> compressors		-	2.08	2.80	2.35
oth auxiliaries		0.05	0.14	0.08	0.10
net electric output		0.06	-1.93	-2.74	-0.61
steam to export	kg/s	4.02	0.25	4.46	3.43
m <sub>CH<sub>4</sub>,eq</sub>	kg/s	2.41	2.88	2.49	2.46
$\eta_{H_2,eq}$	%	<b>81</b>	<b>67</b>	<b>77</b>	<b>75</b>
E <sub>CO<sub>2</sub></sub>	kg <sub>CO<sub>2</sub></sub> /Nm <sup>3</sup> <sub>H<sub>2</sub></sub>	0.82		0.00	0.10
E <sub>CO<sub>2</sub>,eq</sub>	kg <sub>CO<sub>2</sub></sub> /Nm <sup>3</sup> <sub>H<sub>2</sub></sub>	0.76	0.14	-0.04	0.05
Heat rate, HR	Gcal/kNm <sup>3</sup> <sub>H<sub>2</sub></sub>	3.24	0.16	3.28	3.42

An important difference between the CLC+SMR and CLR is the oxygen carrier conversion between the reactors. In the CLC+SMR, the external solids circulation rate and the amount of air used are tuned to reach a limited temperature difference between the AR and FR and to ensure complete fuel conversion to CO<sub>2</sub> and H<sub>2</sub>O. The resulting solids conversion<sup>2</sup> ( $\Delta X_{OC}$ ) is 9.3% and the oxygen carrier is completely oxidized at the AR outlet ensuring always the presence of oxygen in the FR. In case of CLR, the oxygen in the OC is also taking part in the reforming

<sup>2</sup> The solids conversion is defined as the difference in mass of the oxygen carrier between the air and fuel reactor as in the

following equation 
$$\Delta X_{OC} = \frac{m_{(NiO+Ni)_{ox}} - m_{(NiO+Ni)_{red}}}{\max(m_{NiO})}$$

reactions; therefore a sub-stoichiometric amount has to be used. This results in a  $\Delta X_{OC}$  of 5.5% and the oxygen carrier is completely reduced at the fuel reactor outlet.

Table 2: thermodynamic conditions of the two plants shown in Figure 1 and Figure 2.

CHEMICAL LOOPING COMBUSTION + STEAM METHANE REFORMING (Error! Reference source not found.)															
	m	T	P	m·LHV	composition, mol fraction										LHV
	kg/s	°C	bar	MW	C <sub>1</sub>	C <sub>2</sub>	C <sub>3</sub>	C <sub>4</sub>	CO <sub>2</sub>	CO	O <sub>2</sub>	N <sub>2</sub>	H <sub>2</sub>	H <sub>2</sub> O	MJ/kg
A01	2.6	15.0	70.0	122.0	0.89	0.07	0.01		0.02			0.01			46.5
A02	9.0	500.0	33.6	105.0	0.22	0.02			0.01				0.01	0.74	11.7
A03	9.0	620.0	32.6	108.2	0.24				0.03				0.07	0.67	12.1
A04	9.0	900.0	32.3	127.7	0.03				0.05	0.11			0.50	0.31	14.2
A05	6.0	110.5	28.9	125.5	0.04				0.17	0.04			0.75		21.0
A06	0.7	30.0	150.8	89.5									1.00		120.0
A07	7.0	30.0	110						0.98	0.01		0.01			0.1
A08	5.6	212.5	1.2	50.5	0.19	0.01			0.47	0.10		0.01	0.23		9.1
A09	9.3	998.6	1.1						0.54					0.45	0.0
A10	20.1	15.0	1.0								0.21	0.79			0.0
A11	16.4	1101.1	1.1								0.05	0.95			0.0
A12	16.4	100.4	1.1								0.05	0.95			0.0
A13	10.4	485.0	92.0											1.00	0.0
A14	4.5	166.4	6.0											1.00	0.0
A15	347.0	1101.1	1.1						mol. fraction: NiO 20%; MgAl <sub>2</sub> O <sub>4</sub> 80%						
A16	343.2	998.6	1.1						mol. fraction Ni 9%; NiO 11%; MgAl <sub>2</sub> O <sub>4</sub> 80%						
CHEMICAL LOOPING REFORMING (Figure 2)															
	m	T	P	m·LHV	composition, mol fraction										LHV
	kg/s	°C	bar	MW	C <sub>1</sub>	C <sub>2</sub>	C <sub>3</sub>	C <sub>4</sub>	CO <sub>2</sub>	CO	O <sub>2</sub>	N <sub>2</sub>	H <sub>2</sub>	H <sub>2</sub> O	MJ/kg
B01	2.6	15.0	70.0	122.0	0.89	0.07	0.01		0.02			0.01			46.5
B02	8.3	500.0	22.0	120.8	0.28	0.02			0.01				0.01	0.68	14.5
B03	8.3	620.0	21.3	124.1	0.29				0.03	0.00			0.08	0.60	14.9
B04	11.2	900.0	21.3	106.8	0.01				0.08	0.12			0.42	0.37	9.6
B05	8.6	115.6	19.1	103.3	0.01				0.24	0.01			0.65	0.09	12.0
B06	0.7	30.0	149.2	85.9									1.00		120.0
B07	6.1	38.3	110.0						1.00						0.0
B08	0.7	30.0	19.1	16.8	0.09				0.11	0.10		0.02	0.68		23.7
B09	12.3	432.4	21.3								0.21	0.79			0.0
B10	9.4	1020.5	21.1									1.00			0.0
B11	16.6	1066.5	1.2						0.03		0.02	0.86		0.09	0.0
B12	16.6	139.8	1.1						0.03		0.02	0.86		0.09	0.0
B13	4.3	488.7	92.0											1.00	0.0
B14	6.4	488.9	92.0											1.00	0.0
B15	3.4	173.9	6.0											1.00	0.0
B16	236.8	1020.5	21.1						mol. fraction: NiO 9%; Ni 25% MgAl <sub>2</sub> O <sub>4</sub> 66%						
B17	233.9	900.0	21.3						mol. fraction Ni 34%; MgAl <sub>2</sub> O <sub>4</sub> 66%						

### 3.2. Economic assessment

An economic assessment of the different plants has been carried out and compared with benchmark technologies. The total equipment costs of the CLC+SMR and CLR are shown in Figure 3. In the first configuration, the CLC+SMR unit cost represents almost 23% of the TEC and about 20% of the TEC is related to the PSA unit. In the second configuration, the CLR unit is about 25% of the TEC and the MDEA unit for the CO<sub>2</sub> separation represents the higher cost of the plant (more than 26%), resulting in a higher cost of the CLR (+43% than CLC+SMR).

The economic model follows the methodology adopted from the global CCS institute [16]. The TOC is the total overnight cost defined according to NETL [17] which includes any “overnight” capital expenses incurred during the capital expenditure period, except for the escalation and interest during construction (direct costs, indirect cost and contingencies).



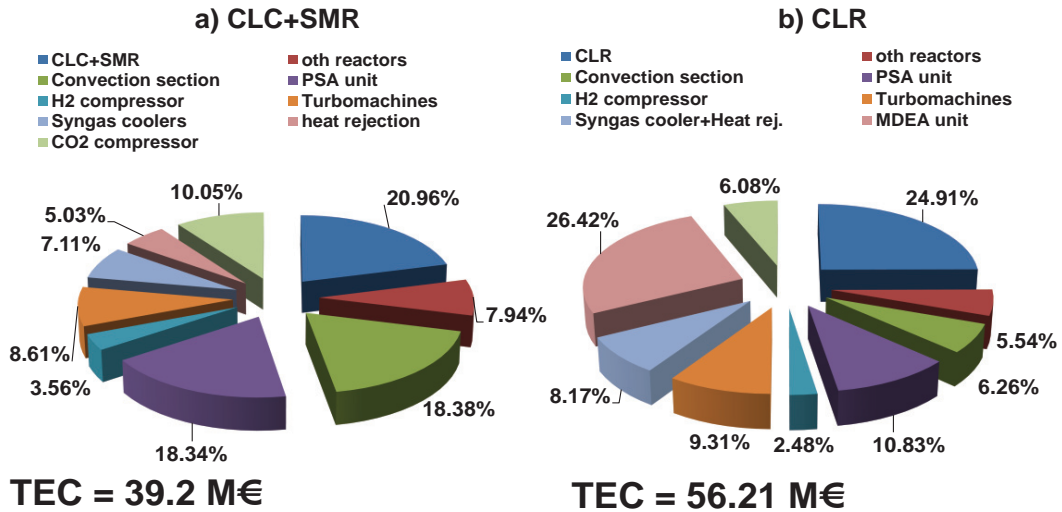


Figure 3: Total equipment cost of the two plant configurations: a) SMR+CLR; b) CLR.

The results of the economic analysis are shown in Table 3. The highest total overnight cost (TOC) is in the case where the CO<sub>2</sub> is separated through chemical absorption. The cost of the CLC+SMR unit is about 30% more expensive than the reformer furnace unit of the conventional SMR. The other costs are mostly the same as for the conventional SMR unit. In case of CLR the cost of the convective section is not as important as in the other cases, however a higher cost of turbomachines is considered due to the presence of the gas turbine. The natural gas fuel consumption represents 50 to 60% of the total cost of hydrogen. The electricity import (except for the conventional unit without CO<sub>2</sub> capture) as well as the steam export slightly affects the final cost of H<sub>2</sub> (COH). The CLC+SMR plant presents a COH about 8% higher than the conventional technology resulting in a cost of CO<sub>2</sub> avoided of 20 €/t<sub>CO2</sub>. In case of CLR, the COH is almost 30% higher than the conventional plant (with a corresponding CO<sub>2</sub> avoidance cost of 85.5 €/t<sub>CO2</sub>). Both plants show better economic performance than the conventional system integrated with a MDEA unit. The lower COH for the CLC+SMR plant compared to the other technologies (integrated with CO<sub>2</sub> capture) is due to the higher H<sub>2</sub> yield as discussed before.

Table 3: Economics of the studied plants.

		SMR	SMR	CLC+SMR	CLR
CO <sub>2</sub> capture		N/A	CA-MDEA		
op. Conditions				900/32	900/20
pure H <sub>2</sub>	kg/s	0.75	0.70	0.75	0.72
	Nm <sup>3</sup> /h	30259	28211	30101	28899
E <sub>CO<sub>2</sub>,eq</sub>	kg <sub>CO<sub>2</sub></sub> /Nm <sup>3</sup> <sub>H<sub>2</sub></sub>	0.76	0.16	-0.04	0.05
Total Overnight Cost	M€	89.34	131.98	92.51	132.63
other O&M fix cost	M€/y	6.29	9.37	6.53	9.58
natural gas	M€/y	31.68	31.68	31.68	31.68
electricity cost	M€/y	-0.02	1.06	1.64	0.37
steam	M€/y	-1.34	-0.09	-0.15	-0.01
total cost	M€/y	50.28	62.22	53.85	61.91
Cost of Hydrogen	€/kg <sub>H<sub>2</sub></sub>	2.36	3.14	2.54	3.05
Cost of Hydrogen	c€/Nm <sup>3</sup> <sub>H<sub>2</sub></sub>	21.08	27.97	22.69	27.17
CO <sub>2</sub> avoidance (equivalent)	€/t <sub>CO<sub>2</sub></sub>	-	114.95	20.10	85.54

### 3.3. Plants feasibility

Based on the techno-economic performance, the CLC+SMR as well as the CLR represent a convenient alternative to the current state-of-the-art technologies when CO<sub>2</sub> capture is considered. Specifically, their feasibility is mostly subjected to the capability of the industry and scientific community to sort out the main challenges associated with these new technologies.

For the CLC+SMR plant, the main challenges are related to the possibility to integrate the tubes filled with the reforming catalyst inside the fuel reactor. Moreover, due to the high temperature required for the reforming, both fuel and air reactors are operated at about 1000 °C and the solids circulation rate is very high to release most of the heat in the fuel reactor. The CLC+SMR is flexible in the use of the oxygen carrier, which represents an important advantage for its development.

For the CLR configuration, the most critical technical challenge is the possibility to operate the interconnected fluidized bed reactors at elevated pressures. In terms of techno-economic performance, the CO<sub>2</sub> avoidance cost is more than 25% lower than conventional technologies due to improved thermal integration.

## 4. Conclusions

Two chemical looping based plants, referred to as CLC+SMR and CLR, for the production of H<sub>2</sub> with CO<sub>2</sub> capture using natural gas as feedstock have been studied and compared with reference technologies from a techno-economic point of view. The optimal reforming operating temperature has been selected at 900 °C while the preferred reforming pressure for the CLC+SMR is 32 bar and for the CLR 20 bar related to the tuning of the pressurized conditions of the interconnected fluidized beds and the gas turbine. The CLC+SMR performance is mostly affected by the CO<sub>2</sub> compression and the lower  $\eta_{H_2,eq}$  is related to the higher electric consumption of the plant. In case of CLR, the resulting equivalent H<sub>2</sub> efficiency is lower because of the lower H<sub>2</sub> yield, while the electric consumption is slightly higher than the benchmark technology without CO<sub>2</sub> capture. Both technologies show a better techno-economic performance than the conventional SMR integrated with a MDEA unit and H<sub>2</sub> firing in the furnace.

The COH associated with CLC+SMR is slightly higher than the conventional system without CO<sub>2</sub> capture and the cost of CO<sub>2</sub> avoided is about 20 €/t<sub>CO2</sub> demonstrating that it is a promising technology for the short mid-term. For the CLR the economic advantages are expected in the mid-long term and in presence of co-production of H<sub>2</sub> and electricity in order to reduce the cost of CO<sub>2</sub> avoided below the current 85 €/t<sub>CO2</sub>.

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