# Techno-economic assessment of chemical looping reforming of Natural Gas for Hydrogen production and power generation with integrated CO<sub>2</sub> capture 3

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# 12 Abstract:

13 The current study presents the techno-economic analysis of the CLR-CC process. The CLR-CC process comprises of chemical looping reforming (CLR) of Natural Gas, water gas shift, CO<sub>2</sub> 14 15 capture and compression, and combined cycle power plant. A 1-D phenomenological model 16 was developed using MATLAB and is used to study the performance of CLR, whereas the 17 remaining part of the process was analysed using commercial software tools like Aspen and Thermoflow. The effect of design conditions in CLR, mainly the air flowrate to the oxidation 18 19 reactor, oxidation reactor outlet temperature and the steam flowrate to the fuel reactor of CLR, 20 on the overall techno-economic performance of the CLR-CC process is reported. The CH4 conversion in CLR, net electrical efficiency, CO<sub>2</sub> avoidance rate and the Levelised Cost of 21 Electricity (LCOE) have been identified as techno-economic performance indicators. For the 22 sensitivity study carried out in this study through 12 cases, the net electrical efficiency of the 23 CLR-CC process varies between 40.0 and 43.4 %, whereas the LCOE varies between 75.3 and 24 144.8 \$/MWh, which is highly dependent on the fuel cost and process contingency rates. 25

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 cycle power plants; Techno-economic analysis.

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42	Nomenclature:
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BEC	Bare Erected Cost
CCS	Carbon Capture and Sequestration
CLC	Chemical Looping Combustion
CLR	Chemical Looping Reforming
CF	Capacity Factor
EPCC	Engineering Procurement and Construction Cost
FC	Fuel Cost
FCF	Fixed Charge Factor
FOM	Fixed Operating and Maintenance
GT	Gas Turbine
HP	High Pressure
HR	Heat Rate
HRSG	Heat Recovery Steam Generator
HTS	High Temperature Shift
LCOE	Levelised Cost of Electricity
LP	Low Pressure
LTS	Low Temperature Shift
MP	Medium Pressure
NG	Natural Gas
ST	Steam Turbine
TCR	Total Capital Requirement
TOC	Total Overnight Cost
TPC	Total Plant Cost
VOM	Variable Operating and Maintenance
WGS	Water Gas Shift
η	Net Electrical Efficiency

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# 46 **1. Introduction**

While the energy transition is taking momentum and a shift towards renewables is evidently 47 visible, oil, coal and natural gas still account for more than 80% of the world's primary energy 48 demand (WEO 2016). There needs to be strike between satisfying the energy demands and the 49 control of CO<sub>2</sub> levels in the atmosphere, as CO<sub>2</sub> is the major contributor to the greenhouse gas 50 emissions. Carbon Capture and Sequestration (CCS) is one of the methods to mitigate 51 greenhouse gas emissions and is foreseen to reduce one sixth of the total CO<sub>2</sub> emissions by 52 2050 (ETP 2012). Three main CO<sub>2</sub> capture routes have been studied and presented in literature, 53 which are pre-, post- and oxy-combustion. A detailed review on the developments in the capture 54 55 methods have been presented by Boot-Handford et al. (2014). The focus of this paper is on a pre-combustion capture method in Natural Gas (NG) based power plants with Chemical 56 Looping Reforming (CLR). 57

58 Chemical Looping (CL) processes with their ability to inherently separate air and  $CO_2$  have 59 attracted a lot of research attention. CL processes like Chemical Looping Combustion (CLC)

and Chemical Looping Reforming (CLR) use metallic oxygen carriers to convert the chemical 60 potential of fossil fuels into work. The concept of chemical looping was first proposed by 61 Richter and Knoche (1983) and was applied to study the CLC based power plant by Ishida, 62 Zheng, and Akehata (1987), Ishida and Jin (1994). CLC completely converts the chemical 63 exergy of fuel into heat at low temperatures (Iloeje, Zhao, and Ghoniem 2015, Naqvi and 64 Bolland 2007, Consonni et al. 2006) whereas CLR converts the chemical exergy of fossil fuel 65 into chemical exergy of hydrogen rich fuel (Nazir, Bolland, and Amini 2017, de Diego et al. 66 2009, Rydén, Lyngfelt, and Mattisson 2006). 67

Figure 1 shows the schematic of a CLR process, which comprises of oxidation reactor and fuel 68 reactor. Compressed air oxidizes the metallic oxygen carrier in the oxidation reactor and 69 70 produces metal oxide and a depleted air stream (N<sub>2</sub>-rich stream). The metal oxide then reacts with NG in the fuel reactor in presence of steam to produce syngas and regenerate the metallic 71 oxygen carrier, which is re-circulated to the oxidation reactor. The current state-of-the-art for 72 CLR is given in a number of studies; on choice of oxygen carrier (Tang, Xu, and Fan 2015, 73 Adanez et al. 2012), reactor scale modeling and experimental studies (Spallina, Gallucci, et al. 74 2016, Francisco Morgado et al. 2016, Diglio et al. 2016, Yahom et al. 2014, Bischi et al. 2012, 75 Pröll et al. 2011, Pröll et al. 2010, de Diego et al. 2009, Rydén, Lyngfelt, and Mattisson 2006). 76

Studies have also been reported on hydrogen production for power generation by Ca-Cu looping

processes (Abanades et al. 2010, Martínez et al. 2014), auto-thermal reforming (Romano,

Processes (Abanades et al. 2010, Martínez et al. 2014), auto-mermai reforming (Romano,
 Chiesa, and Lozza 2010, Nord, Anantharaman, and Bolland 2009, Corradetti and Desideri 2005,

Lozza and Chiesa 2000a, Zohrabian et al. 2016, Ding and Chan 2008, Fiaschi et al. 2005) and

steam-methane reforming (Lozza and Chiesa 2000b, Antzara et al. 2015).

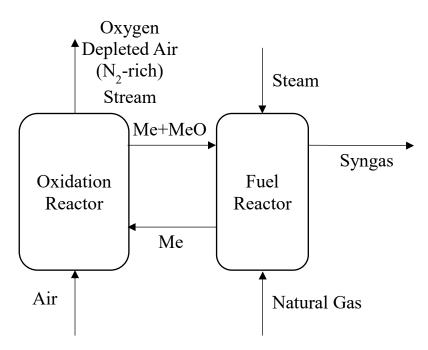


Figure 1: Schematic of a CLR process

82 Analysis of power plants with pre-combustion capture in NG based plants have been presented

by Fan and Zhu (2015), Cormos, Petrescu, and Cormos (2014), Martínez et al. (2013), Cormos

84 (2012), Kvamsdal, Jordal, and Bolland (2007). Techno-economic analysis of combined cycle

with CO<sub>2</sub> capture have been studied by Mathieu and Bolland (2013), Zohrabian et al. (2016),

Spallina, Pandolfo, et al. (2016), Mantripragada and Rubin (2013). The cited literature focuses 86 on pre-combustion methods with hydrogen production through different routes like Ca-Cu 87 looping, steam-methane reforming, auto-thermal reforming and membrane assisted reforming. 88 Anyhow, this paper focuses on the techno-economic analysis of CLR-CC process using the 1-89 D generic phenomenological model for fluidized bed CLR (Francisco Morgado et al. 2016). 90 The CLR-CC process has been defined by Nazir, Bolland, and Amini (2017). The CLR-CC 91 process combines the reforming of NG in CLR, followed by Water Gas Shift (WGS) process, 92 CO<sub>2</sub> capture and compression to produce a H<sub>2</sub>-rich stream, which is used in a combined cycle 93 power plant to produce electricity. The technical performance of the CLR-CC process is studied 94 at different design conditions in the CLR. Net electrical efficiency and CO<sub>2</sub> avoidance rates 95 have been chosen as indicators of technical performance. The effect of air flowrate and 96 temperature at the outlet of oxidation reactor and the steam flow rate in fuel reactor of CLR on 97 98 the techno-economic behavior of the CLR-CC process is shown in this study. The Levelised Cost of Electricity (LCOE) and cost of CO<sub>2</sub> avoidance is estimated for the CLR-CC process. 99 Based on the results, the effect of fuel costs and process contingencies on the LCOE is also 100 presented in this study. The remainder of the sections have the description of the process, the 101 methodology, results and discussions followed by conclusions. 102

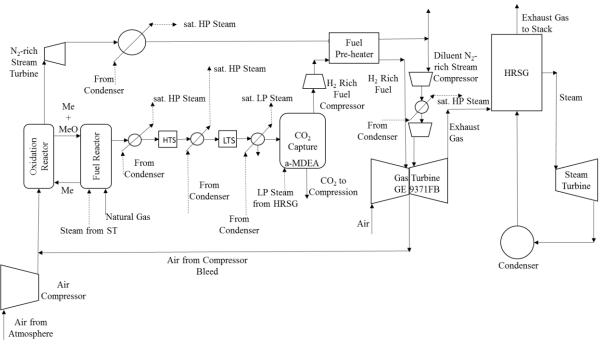
## 103 2. Process Description

Figure 2 shows the schematic of the CLR-CC process. The choice of the design pressure for the 104 CLR and the selection of process systems for the CLR-CC process have been discussed and 105 presented by Nazir, Bolland, and Amini (2018). The design pressure for the CLR is 18 bar. 106 Compressed air at 18 bar reacts with the metallic oxygen carrier in the oxidation reactor. The 107 compressed air is a mixture of air bled from the GT system and the atmospheric air, which is 108 compressed in an additional air compressor. The amount of air bled from the GT system is equal 109 to 12% of the total airflow in the GT. The overall energy penalty is less when the air bled from 110 the GT system is used. Anyhow extracting too much air from the GT system before the 111 combustion chamber might affect the performance and temperature profiles of the GT (Nord, 112 Anantharaman, and Bolland 2009). The metal-metal oxide considered in the current study is 113 Ni-NiO system. A mixture of Ni-NiO leaves the oxidation reactor along with the air stream, 114 which is depleted in Oxygen (N<sub>2</sub>-rich stream). The NiO from the mixture then reacts with NG 115 (100% CH<sub>4</sub> in this study) in the presence of steam in the fuel reactor of CLR. The methane is 116 reformed to syngas and NiO is reduced to Ni. The overall reactions taking place in the CLR 117 unit (reactions 1 and 2) are shown below. 118

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120 $0.5 O_2 + Ni \leftrightarrow NiO$	(1)
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$$121 \qquad \qquad CH_4 + Ni0 \leftrightarrow CO + 2H_2 + Ni \qquad (2)$$





## Figure 2: Schematic of the CLR-CC process

The syngas from the fuel reactor is cooled down and sent through high (HTS) and low 125 temperature (LTS) WGS reactors where most of the CO and H<sub>2</sub>O is converted to CO<sub>2</sub> and H<sub>2</sub>. 126 Syngas and the HTS product stream are cooled down to produce saturated High Pressure (HP) 127 steam at 174.4 bar. The pressure at which steam is produced is dependent on the point it is being 128 129 integrated within the steam cycle. The product stream from LTS is cooled down to 50 °C. 130 Saturated Low Pressure (LP) steam at 3.8 bar is produced from cooling of LTS product. The final gaseous mixture contains mainly CO<sub>2</sub> and H<sub>2</sub> and is ready for CO<sub>2</sub> capture. CO<sub>2</sub> is 131 absorbed in the absorber using the a-MDEA amine and H<sub>2</sub>-rich fuel is collected at the top. The 132 rich amine solution is then flashed and pre-heated before entering the regenerator. The amine 133 is regenerated and is sent to the absorber, whereas the CO<sub>2</sub> stream is compressed and prepared 134 135 for storage.

136 The H<sub>2</sub>-rich fuel from the top of the absorber is compressed, preheated and sent to the Gas Turbine (GT) for combustion with air. 12% of the compressed air in the GT, about 277 TPH 137 138 from each GT system, is extracted as bleed from the compressor discharge and is used in the oxidation reactor of CLR. The N2-rich stream from the oxidation reactor is expanded in a 139 turbine to extract work and then is cooled down by producing saturated HP steam at 174.4 bar 140 and pre-heating the H<sub>2</sub>-rich fuel. Fraction of the N<sub>2</sub>-rich stream, equal to the mass of air bled 141 from the GT, is compressed and used as a diluent in the GT. The inter-stage cooling during 142 compression of N<sub>2</sub>-rich stream is also used to produce saturated HP steam at 174.4 bar. Similar 143 approach to treat the N<sub>2</sub>-rich stream has been followed in Nazir, Bolland, and Amini (2018). 144 Several other process alternatives were considered by the authors to treat the N<sub>2</sub>-rich stream. 145 For example, cooling the fraction of N<sub>2</sub>-rich stream from the oxidation reactor, which is used 146 as a diluent in the GT system, and compressing it to the desired pressure in the GT. Anyhow, 147 the authors noticed that treating the N<sub>2</sub>-rich stream as presented in this study has less efficiency 148

149 penalty on the overall process.

The power plant is a combined cycle with two gas turbines, two Heat Recovery Steam 150 Generators (HRSG) and one steam turbine (ST) system, as it is the same configuration used for 151 a NGCC plant without capture in EBTF (2011). The steam cycle is a three-pressure level with 152 reheat and comprises of one high pressure steam turbine, one medium pressure steam turbine 153 and two flow low pressure turbines. The low pressure (LP), medium pressure (MP) and high 154 pressure (HP) steam levels are maintained at 3.4, 32.7 and 166 bar, respectively. The water and 155 steam mixture from the ST is condensed in a water-cooled condenser before the water is 156 pumped and sent to HRSG. The cooling water requirements in the process is met by a natural 157 draft cooling tower. The saturated HP and LP steam generated from cooling of process streams 158 in the process is added to the HRSG at the inlet of respective HP and LP superheaters. The 159 assumptions made in the model are explained in the following section. 160

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# 162 **3. Methodology**

163 The techno-economic assessment of the CLR-CC process was carried out using the process 164 models to assess different sections of the process, and the economic model as described by 165 GCCSI (2013). The description of the models and the respective assumptions alongside criteria 166 for technical assessment is briefed below.

167 3.1. 1-D Model for CLR

The 1-D model used in this work consists of a 1-D generic phenomenological model for 168 fluidized bed reactors applied to CLR (Morgado et al. 2016) developed using MATLAB. The 169 generic model formulation is based on the averaging probabilistic approach developed by 170 Thompson et al. (Thompson, Bi, and Grace 1999, Abba et al. 2003) and couples the three most 171 frequent fluidization regimes in industry (bubbling, turbulent and fast fluidization). 172 Furthermore, it relies on the two-phase theory that distinguishes between a low and high dense 173 phase, poor and highly concentrated in solids, respectively. The material and energy balances 174 as well as the empirical closure laws used to describe the hydrodynamics of the system under 175 different fluidization regimes are described by Morgado et al. (2016). The use of kinetic models 176 like this one helps in evaluating the process more accurately at different design conditions and 177 dynamics of the process. In this work the Dual Circulating Fluidized Bed (DCFB) configuration 178 proposed by (Pröll et al.) was considered. Therefore, both reactors operate under the same 179 fluidization regimes that is turbulent and/or fast fluidization (Schmid et al. 2011, Kolbitsch et 180 al. 2009). 181

Adiabatic conditions were assumed in both oxidation and fuel reactors. The temperature at the 182 183 outlet of the oxidation reactor was limited to  $1200 \pm 10$  °C due to the thermal degradation of the oxygen carrier and was used to estimate the oxygen carrier circulation rate between the 184 oxidation and fuel reactors. In addition, the effect of changing the temperature at the outlet of 185 oxidation reactor to  $1100 \pm 10$  °C is also presented in this paper. The air flowrate entering the 186 oxidation reactor was defined to meet higher conversion rates of methane in the fuel reactor. 187 The amount of steam flowrate fed to the fuel reactor was assumed based on the CO/H<sub>2</sub>O ratio 188 required for favorable conditions in WGS. 189

190 The dimensions of the reactors (height and diameter) were established in order to meet the 191 equilibrium conversions in the fuel reactor alongside maintaining the fluidization regimes as in 192 DCFB. Due to the excellent heat transfer properties of fluidized bed reactors, the temperature in the low and high dense phases was considered equal. The superficial velocity of the gas
inside the reactors has been constrained so that it is always higher or equal to the minimum
fluidization velocity. The particle size of the oxygen carrier is assumed 250 µm.

196 3.2. WGS and CO<sub>2</sub> capture model

The WGS reactors, CO<sub>2</sub> capture and compression processes were simulated in ASPEN Hysys 197 V8.6 (AspenHYSYS 2017). Peng-Robinson thermodynamic model is considered for the WGS 198 and CO<sub>2</sub> compression sections, whereas Acid-Gas Model is used to estimate the equilibrium 199 conditions in CO<sub>2</sub> capture section. The HTS and LTS are modeled using steady state 200 equilibrium reactor modules with adiabatic conditions. The inlet streams to the HTS and LTS 201 reactors are at 400 °C and 200 °C respectively. The pressure drop in the WGS reactors is 202 assumed 3%. The pressure drop considered in the heat exchangers in the entire process is 2% 203 for gaseous streams and 0.4 bar for liquid streams (EBTF 2011). 204

The main design conditions in the  $CO_2$  capture section are listed in Table 1. The amine used to 205 absorb CO<sub>2</sub> is a-MDEA (45% by mass in the solution), which is used for moderate partial 206 pressures of CO<sub>2</sub> (3-4 bar) at the absorber inlet (Nord, Anantharaman, and Bolland 2009) and 207 5% by mass Piperazine is used as an activator. The capture rate of 95% is assumed across the 208 absorber and the flowrate of amine is estimated. Superheated LP steam extracted from the inlet 209 of the LP steam turbine at 3.4 bar and 270 °C is used in the reboiler of the regenerator. CO2 210 captured is compressed and pumped to 110 bar in three compression stages followed by 211 pumping as described in EBTF (2011). 212

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Number of absorber trays	20
Number of stripper trays	20
Pressure drop in the absorber (bar)	0.1
Pressure drop in the regenerator (bar)	0.1
Lean amine loading (mol CO2/mol MDEA)	0.301
MDEA/water (mass/mass)	1
Condenser Temperature in regenerator (°C)	46.11
Adiabatic efficiency of pump for regenerated amine (%)	80

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*Table 1: Design conditions in CO*<sub>2</sub> *capture section (Nazir, Bolland, and Amini* 2017)

# 215 3.3. Power Plant

The combined cycle power plant has been analysed using Thermoflex component of the 216 Thermoflow Suite (Thermoflow 2017). The GT system chosen for the analysis is GE-9371FB, 217 which is robust to changes in fuel composition and is favorable for H<sub>2</sub>-rich fuels (EBTF 2011, 218 219 Nord, Anantharaman, and Bolland 2009). The power plant comprises of two GTs, two HRSGs and one ST system. The ST system is a three steam level with reheat. The steam levels are 220 221 3.4/32.7/166 bar. The GT is run at full load conditions for all the cases considered in this paper 222 and hence the fuel input to the GT is estimated accordingly. 12% of the compressed air is bled at the compressor discharge in the GT and used in the CLR oxidation reactor. The N<sub>2</sub>-rich 223 224 stream from the fuel reactor of CLR is added in the combustor along with the fuel not only to 225 compensate for the mass of air bled from the GT system, but also to act as a diluent which 226 reduces the flame temperature when H<sub>2</sub>-rich fuel is combusted (Chiesa, Lozza, and Mazzocchi 227 2005).

## 228 3.4. Economic Model

The LCOE and the cost of  $CO_2$  avoidance are the main performance indicators for the economic analysis of the process. The economic analysis to assess the LCOE and cost of  $CO_2$  avoidance for the process is carried out using the methodology proposed by the GCCSI (2013). The LCOE for the CLR-CC process is estimated using the following equation 1:

233 
$$LCOE = \frac{(TCR)(FCF) + FOM}{(MW)(CF \times 8766)} + VOM + (HR)(FC)$$
Equation (1)

# The nomenclature used in equation 1 is given in Table 2.

Parameter	Definition	Unit
TCR	Total Capital Requirement in the base year of the analysis	\$
FCF	Fixed Charge Factor as defined in equation 2	fraction
FOM	Fixed O&M costs	\$/year
MW	Net power output of the plant	MW
CF	Capacity Factor – availability of the plant	Fraction
VOM	Variable O&M costs excluding the fuel costs	\$/MWh
HR	Net power plant heat rate	MJ/MWh
FC	Fuel Cost per unit of energy	\$/MJ

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Table 2: Nomenclature for parameters used to estimate LCOE in equation 1

The FCF is calculated using equation (2) where "r" is the interest rate or discount rate and T is

the economic life of the plant relative to the base year of analysis used in the study. Furthermore,

an interest rate of 10% and an economic life of the plant of 30 years were assumed.

239 
$$FCF = \frac{r(1+r)^T}{(1+r)^{T-1}}$$
 Equation (2)

## 240 The TCR is estimated using the methodology as shown in Table 3.

Component	Definition
Bare Erected Cost (BEC)	Sum of installed cost of equipment
Engineering Procurement Construction Costs (EPCC)	10% of BEC
Process Contingency	40%+ of BEC
Project Contingency	15 - 30 % of (BEC +EPCC + Process Contingency)
Total Contingencies	Process Contingency + Project Contingency
Total Plant Costs (TPC)	BEC +EPCC + Total Contingencies
Owners Cost	20.2% of TPC (NETL 2011)
Total Overnight Costs (TOC)	TPC + Owners Cost
Total Capital Requirement (TCR)	1.14*TOC (NETL 2011)

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Table 3: Methodology to estimate TCR

The Sizing and Economics tool in ASPEN Hysys V8.6 and the PEACE component in Thermoflow V26 is used to estimate the installation costs of the process equipment except the oxidation and fuel reactors of CLR. The LCOE for the NGCC without capture case estimated using the BEC from the database of commercial software tools, like Aspen Hysys and Thermoflow, is validated against the LCOE reported in the DOE/NETL (2007) considering

similar fuel costs. The costs of the basic equipment considered in this paper are from the 247 reference year 2016, and hence, correction factors have not been introduced. The BEC of high 248 temperature and high pressure reactors is difficult to estimate and the cost data is not readily 249 available. Hence, the methodology described in Peters and Timmerhaus (1991) is used to 250 estimate the cost of the oxidation and fuel reactors of CLR, where the weight of the reactor is 251 calculated first. The height and diameter of the oxidation and fuel reactors were considered 6 252 m and 6 m, respectively since the equilibrium conditions are reached within those dimensions. 253 The weight of each reactor is calculated to be 364750 lb. A reference cost of the reactor similar 254 to that of Fluidized Catalytic Cracker is used in this study (Spallina, Pandolfo, et al. 2016). The 255 reference cost assumed is 8.2 M\$ for 130000 lb. With a scale factor of 0.6, the cost of each 256 reactor is 15.23 M\$. Considering installation cost to be 80% of the cost of the reactor, the BEC 257 for each reactor is 27.4 M\$. The transport disengaging height (TDH) in the reactor is not 258 259 considered in this study, since the height of the reactor assumed is enough to reach equilibrium conditions. Considering TDH, i.e. assuming 20 m height of the reactor instead of 6 m in this 260 study will increase the BEC of the CLR by 30% but has less than 1% effect on the LCOE of 261 the process. 262

As seen in Table 3, the process contingency is 40%+ of the BEC as the process is a new concept with limited data. However, in this study, the process contingency is assumed 50% of BEC for the CLR-CC process. On the other hand, a NGCC plant without capture will have a process contingency of 10% of BEC, as it is already a commercially available technology. A project contingency of 30% of sum of BEC, EPCC and process contingency is assumed in this study for all the cases. The TCR/TOC ratio of 1.14 is assumed for the CLR-CC process as the project is assumed a high-risk investor owned utility (NETL 2011).

The assumptions made to estimate the operating and maintenance (O&M) costs is shown inTable 4.

Fixed O&M Costs		
Operating Labor	1.7	M\$
Maintenance, Support and Administrative Labor	2.5	% of TOC
Property Taxes	Included in insurance costs	
Insurance costs	2	% of TOC
Cost of NG (Fuel Cost)	10.18	\$/GJ LHV
Variable O&M Costs		
Consumables		
Cooling Water Make Up Costs	0.39	\$/m3
Process Water Cost	2.22	\$/m3
Catalysts and Sorbent Replacement		
Oxygen Carrier cost	15	\$/kg
WGS catalyst cost	15574	\$/m3
Amine cost	2298.3	\$/m3
Replacement Period	5	Years
CO2 Transport and Storage Costs	11.12	\$/ton CO2
Emissions Tax (CO <sub>2</sub> tax)	27.22	\$/ton CO <sub>2</sub>

After estimating the LCOE of the CLR-CC process, the cost of CO<sub>2</sub> avoided is estimated by 273 equation (3). 274

 $Cost of CO_2 avoided \left(\frac{\$}{tCO_2}\right) = \frac{LCOE_{CLR\_CC} - LCOE_{NGCC}}{\left(\frac{tCO_2}{MWh}\right)_{NGCC} - \left(\frac{tCO_2}{MWh}\right)_{CLR\_CC}} \quad \text{Equation (3)}$ 276

#### 277 3.5. Criteria for techno-economic assessment

278 The performance of CLR affects the overall performance of the CLR-CC process. The conditions of pressure, temperature and compositions of the product streams from the CLR 279 affect the fuel flowrates in the process, the turbines and compressor work, and amount of steam 280 produced from the cooling of high temperature process streams. The available manipulative 281 variables in the process are the air flowrate (O<sub>2</sub> flowrate) to the oxidation reactor, the outlet 282 temperature of the oxidation reactor, the amount of steam added in the fuel reactor and the 283 design pressure in the oxidation reactor. The impact of pressure inside the oxidation reactor is 284 not included in this work and it forms a part of another article. Thus, only the remaining three 285 independent variables (air flowrate, oxidation reactor outlet temperature and amount of steam 286 to the fuel reactor) were studied in this work. To evaluate the performance of the CLR-CC 287 process while manipulating these independent variables, different performance indicators were 288 defined. The conversion of CH<sub>4</sub> and the oxygen carrier utilization are the main performance 289 indicators for the CLR process. The CO<sub>2</sub> avoidance and the net electrical efficiency are the 290 performance indicators considered for the CLR-CC process. The LCOE is the main 291 performance indicator for the economic performance of the process. The CO<sub>2</sub> avoidance and 292 net electrical efficiency are defined as follows: 293

$$\frac{CO_2 Avoidance (\%) =}{\frac{100 \times (CO_2 \text{ emitted in NGCC without capture} - CO_2 \text{ emitted in NGCC with capture})}{CO_2 \text{ emitted in NGCC without capture}}$$

Equation (4)

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297 Net Electrical Efficiency 
$$(\eta) = \frac{100 \times Net \ electricity \ produced \ from \ the \ overall \ process}{LHV \ of \ NG \ input \ to \ the \ process}$$
 Equation (5)

298

299 Considering the amount of air flowrate to the oxidation reactor, the stoichiometry given by the 300 reforming reaction of CH<sub>4</sub> implies that 0.5 moles of O<sub>2</sub> are needed to reform CH<sub>4</sub> into CO and 301 H<sub>2</sub> (Reaction 3).

(3)

302 
$$CH_4 + 0.5 O_2 \rightarrow CO + 2 H_2$$

Hence, the availability of oxygen in the fuel reactor through the metal oxide (NiO) plays an 303 important role in the conversion of CH4. A sensitivity study was carried out varying the amount 304 305 of oxygen entering the CLR by considering the stoichiometric molar ratio of O<sub>2</sub>/CH<sub>4</sub> in the system to be 0.5, 0.75 and 0.9. In these cases, the temperature at the outlet of oxidation reactor 306 was assumed to be  $1200 \pm 10$  °C and the steam/CH<sub>4</sub> ratio by mass was assumed to be 1. The 307 equilibrium conversion of CH<sub>4</sub> at different O<sub>2</sub>/CH<sub>4</sub> molar ratio is shown in Table 5. 308

309

O <sub>2</sub> :CH <sub>4</sub> (mol/mol)	Conversion of methane (%)
0.5	50.6
0.75	81.9
0.9	96.2
11 5 6	

Table 5: Sensitivity study to decide the  $O_2/CH_4$  ratio

As seen in Table 5, the conversion of  $CH_4$  increases with an increase in  $O_2/CH_4$  ratio at the inlet 311 of the CLR. Hence, further sensitivity studies in this paper have been reported with an O<sub>2</sub>/CH<sub>4</sub> 312 molar ratio of 0.8 and 0.9, where the conversion of CH<sub>4</sub> in the fuel reactor is more than 90%. 313 The overall techno-economic performance of the system was assessed for O<sub>2</sub>/CH<sub>4</sub> ratios of 0.8 314 and 0.9, steam/CH<sub>4</sub> ratio by mass of 0.5, 1 and 1.5 and using oxidation reactor outlet 315 temperatures equal to 1200 °C and 1100 °C. The different cases studied within this work are 316 defined in Table 6. The N<sub>2</sub>-rich stream temperature at the inlet of the N<sub>2</sub>-rich stream turbine is 317 318 same as the oxidation reactor outlet temperature. Although the temperature at the inlet of the 319 N<sub>2</sub>-rich stream turbine is very high in this analysis where the focus is on identifying the potential of the process, it should be noted that in real case scenario, the temperatures are limited by the 320 321 maximum allowable temperature suitable for the rotor blades in the turbine. The amount of CH<sub>4</sub> 322 flow to the fuel reactor is based on matching the amount of H<sub>2</sub>-rich fuel required to maintain a constant 1.55 GW LHV at the inlet of GT system. Any excess H<sub>2</sub>-rich stream produced from 323 324 the reforming process is also reported.

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Cases	O2/CH4 by	Steam/CH <sub>4</sub>	Oxidation Reactor Outlet	CH <sub>4</sub> flow
	moles	by mass	Temperature (°C)	(TPH)
1	0.9	0.5	1200	170
2	0.9	1	1200	170
3	0.9	1.5	1200	172
4	0.9	0.5	1100	170
5	0.9	1	1100	170
6	0.9	1.5	1100	170
7	0.8	0.5	1200	160
8	0.8	1	1200	160
9	0.8	1.5	1200	160
10	0.8	0.5	1100	160
11	0.8	1	1100	160
12	0.8	1.5	1100	160



*Table 6: Definition of cases for techno-economic analysis* 

328

## 329 4. Results and discussion

The main results of the techno-economic analysis of the CLR-CC process for the cases defined in Table 6 are shown in Table 7 and Table 8. Table 7 presents the conditions and results in the CLR at different design conditions with respect to air flowrate ( $O_2/CH_4$  mole ratio), oxidation reactor outlet temperature and steam flowrate in the fuel reactor. Table 8 presents the results for the overall process behavior. The '+' and '-' signs in Table 8 indicates whether the components in the process add or negate the net electrical efficiency respectively. The discussion on these results is presented in this section.

Cases		1	2	3	4	5	6	7	8	9	10	11	12
Oxidation reactor													
Outlet temperature	°C	1200	1200	1200	1105	1100	1100	1200	1200	1200	1100	1100	1100
Outlet pressure	bar	17.94	17.96	17.94	17.93	17.93	17.93	17.80	17.81	17.81	17.74	17.74	17.75
Oxygen carrier flowrate	TPH	12289	9291	7925	22660	18612	13367	6968	6096	5566	11860	9443	8189
N <sub>2</sub> -rich stream flowrate	TPH	1005	1006	1017	1005	1005	1005	841	841	841	841	841	841
Fuel Reactor													
Outlet temperature	°C	973	902	843	977	943	882	864	816	778	894	841	801
Outlet pressure	bar	17.50	17.87	17.68	17.50	17.59	17.69	17.43	17.46	17.51	17.45	17.48	17.54
Syngas flowrate	TPH	560	644	739	560	645	730	495	575	655	495	575	655
Methane conversion	%	98.9	96.6	95.3	98.9	98.8	97.5	91.0	88.3	85.9	94.0	91.5	89.2

Table 7: Design conditions and results from CLR

Cases	Units	1	2	3	4	5	6	7	8	9	10	11	12
Gas Turbine	%-LHV	+25.8	+25.8	+25.4	+25.8	+25.8	+25.8	+27.2	+27.2	+27.2	+27.3	+27.3	+27.3
Steam Turbine	%-LHV	+18.4	+17.4	+16.4	+18.4	+17.8	+17.0	+18.6	+17.6	+16.9	+18.6	+17.8	+17.1
N <sub>2</sub> -rich Stream Turbine	%-LHV	+9.6	+9.6	+9.7	+9.0	+9.0	+9.0	+8.5	+8.5	+8.5	+8.0	+8.0	+8.0
Diluent N2 Stream Compressor	%-LHV	-4.1	-4.1	-4.1	-4.2	-4.2	-4.2	-4.4	-4.4	-4.4	-4.4	-4.4	-4.4
H <sub>2</sub> rich fuel Compressor	%-LHV	-0.8	-0.7	-0.7	-0.7	-0.8	-0.8	-0.8	-0.7	-0.7	-0.8	-0.8	-0.8
Air Compressor	%-LHV	-3.8	-3.8	-3.8	-3.8	-3.8	-3.8	-2.9	-2.9	-2.9	-2.9	-2.9	-2.9
Pump for Regenerated Amine	%-LHV	-0.1	-0.1	-0.1	-0.1	-0.1	-0.1	-0.1	-0.1	-0.1	-0.1	-0.1	-0.1
CO <sub>2</sub> Compressors and Pump	%-LHV	-1.7	-1.8	-1.8	-1.7	-1.8	-1.8	-1.6	-1.6	-1.6	-1.6	-1.7	-1.7
Auxiliaries	%-LHV	-1.1	-1.1	-1.0	-1.1	-1.1	-1.1	-1.1	-1.1	-1.1	-1.1	-1.1	-1.1
Net LHV Input	MW	2363	2363	2391	2363	2363	2363	2224	2224	2224	2224	2224	2224
Net Electrical Efficiency	%	42.2	41.2	40.0	41.6	40.8	40.0	43.4	42.5	41.8	43.0	42.1	41.4
CO <sub>2</sub> Avoidance	%	75.9	82.5	82.6	75.9	84.4	86.1	67.9	72.9	71.3	68.8	75.9	75.5
CO <sub>2</sub> Capture	%	84.6	88.9	89.0	84.6	90.1	91.1	78.2	81.6	80.5	78.8	83.7	83.4
Heat required in stripper reboiler	MJ/kg CO <sub>2</sub>	1.7	1.6	1.6	1.7	1.6	1.6	1.7	1.6	1.6	1.7	1.7	1.6
H <sub>2</sub> -rich fuel at GT inlet	TPH	95.7	80.8	78.6	95.8	81.7	75.9	95.6	82.5	81.6	99.1	83.3	80.4
Excess H <sub>2</sub> -rich stream flow	TPH	1.4	0.5	0.7	1.5	0.8	0.3	2.0	0.7	0.1	3.0	1.4	0.8
Economic Analysis													
TCR	M\$	2097	2060	2050	2070	2080	2069	1944	1937	1922	1932	1924	1918
LCOE	\$/MWh	137.6	138.8	141.9	143.1	144.1	144.8	131.7	134.0	135.9	134.5	136.3	138.3
Cost of CO2 avoidance	\$/tCO <sub>2</sub>	185.7	177.4	188.7	206.8	192.3	191.9	181.8	180.9	193.6	191.9	183.5	193.2

Table 8: Main results from techno-economic analysis of CLR-CC process

## 340 4.1.Behavior of the CLR

Error! Reference source not found. Figure 3, Figure 4 and Figure 5 show the main results for 341 the performance of the CLR at different design conditions described in Table 6. Error! 342 **Reference source not found.** Figure 3, Figure 4 and Figure 5 shows the conversion of CH<sub>4</sub> in 343 the fuel reactor, the oxygen carrier flowrate in the oxidation reactor of the CLR and the syngas 344 temperature at different air flowrates (O<sub>2</sub>/CH<sub>4</sub>), oxidation reactor outlet temperatures and 345 steam/CH<sub>4</sub> ratios (by mass) respectively. Table 9 shows the composition of the syngas for the 346 12 cases defined in Table 6. The conversion of CH<sub>4</sub> in the fuel reactor of the CLR is a function 347 of air flowrate (O<sub>2</sub>/CH<sub>4</sub> mole ratio) in the oxidation reactor, the syngas temperature and the 348 steam flowrate (steam/CH<sub>4</sub> mass ratio) in the fuel reactor. The syngas temperature anyhow is 349 mainly dependent on the steam flowrate (steam/CH<sub>4</sub> mass ratio). The CH<sub>4</sub> conversion in the 350 CLR is higher by 7-10% when the  $O_2/CH_4$  mole ratio in the oxidation reactor is 0.9 when 351 compared to 0.8. The CH<sub>4</sub> conversion is 3-5% higher when the steam/CH<sub>4</sub> mass ratio in the fuel 352 reactor is 0.5 and decreases when the ratio is increased to 1 and 1.5. Higher steam flowrates in 353 the fuel reactor lowers the overall fuel reactor temperature (reflected in the syngas temperature) 354 and hence lowering the CH<sub>4</sub> conversion. The CH<sub>4</sub> conversion is 2- 4% higher when the 355 oxidation reactor outlet temperature is changed from 1200 to 1100 °C. There is significant 356 change in the oxygen carrier usage in the CLR when the oxidation reactor outlet temperature is 357 changed from 1200 to 1100 °C. Lower the oxidation reactor outlet temperature, higher is the 358 oxygen carrier circulation to maintain a steady process. The internal behavior of the oxidation 359 and fuel reactor of the CLR in terms of the average gas and solid axial velocities, average void 360 361 fractions and type of fluidization regimes is discussed further in the section.

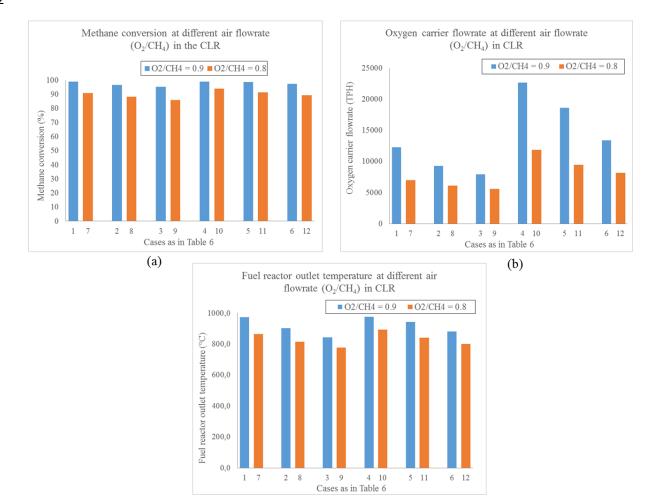
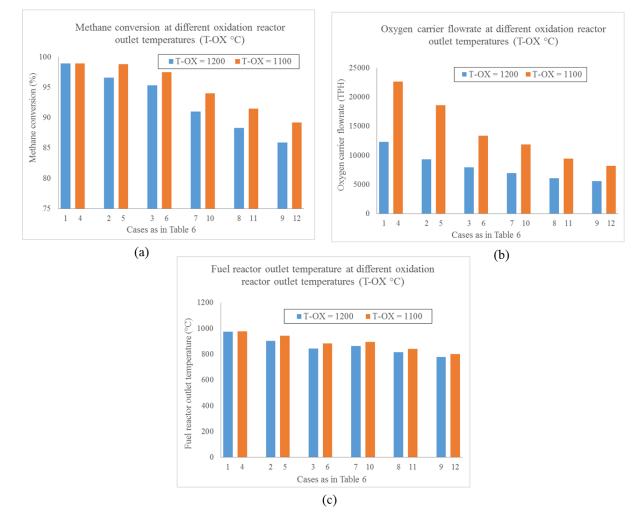


Figure 3: For different air flowrates (O<sub>2</sub>/CH<sub>4</sub>) (a) Methane conversion in the fuel reactor of CLR (b) Oxygen
 carrier flowrate in the CLR (c) Fuel reactor outlet temperature (syngas temperature)

(c)



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Figure 4: For different oxidation reactor outlet temperature (T-OX) (a) Methane conversion in the fuel reactor
 of CLR (b) Oxygen carrier flowrate in the CLR (c) Fuel reactor outlet temperature (syngas temperature)

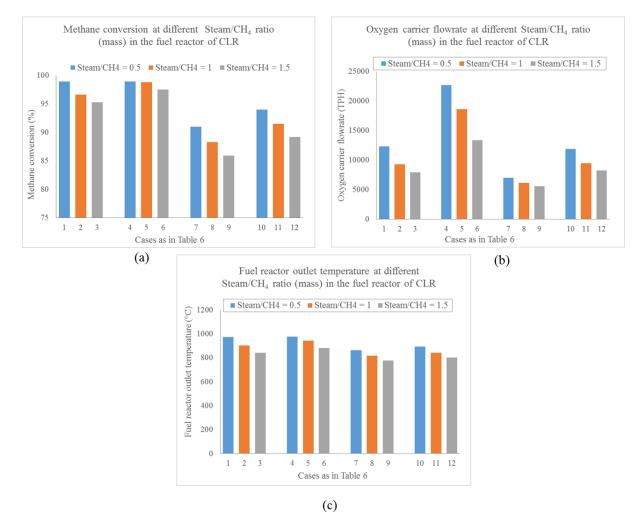
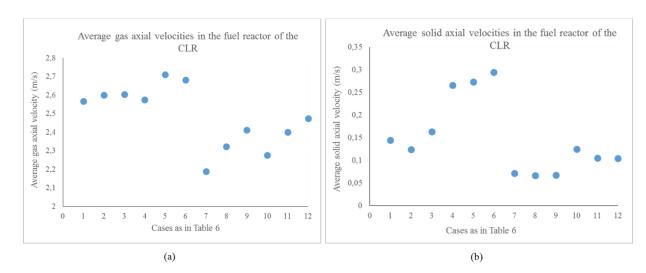


Figure 5: For different steam/CH<sub>4</sub> ratio (mass) in the fuel reactor of the CLR (a) Methane conversion in the fuel
 reactor of CLR (b) Oxygen carrier flowrate in the CLR (c) Fuel reactor outlet temperature (syngas temperature)

	O <sub>2</sub> /CH <sub>4</sub> by	Steam/CH4	Oxidation Reactor Outlet	Syngas composition (mol%)						
Cases	moles	by mass	Temperature (°C)	$CH_4$	СО	$CO_2$	$H_2$	$H_2O$		
1	0.9	0.5	1200	0.3	20.5	8.4	42.5	28.3		
2	0.9	1.0	1200	0.9	13.1	12.1	41.1	32.7		
3	0.9	1.5	1200	1.1	10.8	11.7	36.6	39.8		
4	0.9	0.5	1100	0.3	20.5	8.4	42.5	28.3		
5	0.9	1.0	1100	0.3	15.9	9.7	39.8	34.4		
6	0.9	1.5	1100	0.6	11.7	11.1	37.3	39.4		
7	0.8	0.5	1200	2.7	18.7	9.2	43.8	25.5		
8	0.8	1.0	1200	3.2	13.1	11.0	39.7	33.0		
9	0.8	1.5	1200	3.5	9.3	11.9	36.0	39.3		
10	0.8	0.5	1100	1.8	19.9	8.4	45.1	24.8		
11	0.8	1.0	1100	2.3	14.3	10.3	41.1	32.1		
12	0.8	1.5	1100	2.6	10.3	11.3	37.4	38.3		

Table 9: Composition of syngas at the outlet of the fuel reactor of CLR

- From Figure 6, it is observed that the average gas axial velocities change little with the change
- in the steam flowrate in the fuel reactor for the first 6 cases ( $O_2/CH_4$  mole ratio = 0.9) which is
- about 1.4% in cases 1-3 and about 5% in cases 4-6. For the cases with  $O_2/CH_4$  mole ratio as
- 0.8, a change in the steam/CH<sub>4</sub> mass ratio from 0.5 to 1.5 led to an increase in the average gas axial velocities by 9%. The average gas axial velocities are in the order of 2.62 m/s for O<sub>2</sub>/CH<sub>4</sub>
- mole ratio of 0.9 and are lower for the cases with  $O_2/CH_4$  mole ratio of 0.8. This decrease in
- the velocity with respect to the  $O_2/CH_4$  mole ratio is due to the decrease in the methane flowrate
- 382 required to maintain a steady power production at full load through the CLR-CC process. The
- average axial gas velocity increases by about 3% for the cases with oxidation reactor outlet
- temperature of 1100  $^{\circ}$ C when compared to the cases with 1200  $^{\circ}$ C.
- The average solid axial velocity in the fuel reactor of the CLR for the different cases is shown in Figure 6. The solids axial velocity is affected by the change in the  $O_2/CH_4$  mole ratio. A decrease in the  $O_2/CH_4$  mole ratio from 0.9 to 0.8, halves the average solids axial velocity in the fuel reactor of the reactor. This behavior can be explained by the lower requirements of methane in the fuel reactor. Changes in the steam flow rate do not affect the average solids axial velocity significantly. However, for lower oxidation reactor outlet temperatures, higher axial
- 391 solids velocity is observed due to higher oxygen carrier circulation.
- The average void fraction in the fuel reactor of the CLR is not sensitive to the oxidation reactor 392 outlet temperature as seen in Figure 7. However, it is affected by the steam flowrate in the fuel 393 reactor and O<sub>2</sub>/CH<sub>4</sub> mole ratio in the oxidation reactor of the CLR. An increase in steam 394 flowrates in the fuel reactor results in an increase in the fast fluidization regime contribution 395 and consequently higher the average void fractions in the fuel reactor. An increase of the 396 steam/CH<sub>4</sub> mass ratio in the fuel reactor from 0.5 to 1.5 with O<sub>2</sub>/CH<sub>4</sub> mole ratio of 0.9 leads to 397 an increase in the void fraction by 12%. For the cases with O<sub>2</sub>/CH<sub>4</sub> mole ratio of 0.8, an increase 398 of the steam/CH<sub>4</sub> mass ratio in the fuel reactor from 0.5 to 1.5 increases the average void 399 fraction by 6% in the fuel reactor. 400
- 401



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403 Figure 6: (a) Average gas axial velocity along the bed of the fuel reactor of the CLR (b) Average solid axial
 404 velocity along the bed of the fuel reactor of the CLR

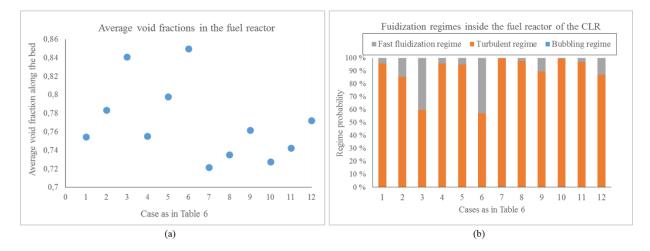




Figure 7: (a) Average void fraction in the fuel reactor of the CLR (b) Fluidization regime probabilities in the fuel reactor of the CLR

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Figure 8 shows the average gas axial velocity in the oxidation reactor of the CLR. Due to lower air flowrates for cases with  $O_2/CH_4$  mole ratio of 0.8, the average axial velocity of the gas decreases by 7-9% in the oxidation reactor when compared to cases with the  $O_2/CH_4$  mole ratio of 0.9. Reducing the oxidation reactor outlet temperature from 1200 to 1100 °C leads to a decrease in the average gas axial velocities by 4.4% due to an increase in the gas density.

415 A decrease in the  $O_2/CH_4$  mole ratio from 0.9 to 0.8 halves the average solids axial velocity in 416 the oxidation reactor as seen in Figure 8. This is because of the lower air flowrates in the 417 oxidation reactor when the methane requirements are low in the fuel reactor. It is also reflected 418 in having higher contribution of the turbulent fluidization regime as seen in Figure 9. Cases 1-419 6 operate mostly under fast fluidization regime. Hence, the average void fraction in the oxidation reactor is 0.97 when the  $O_2/CH_4$  mole ratio is 0.9, whereas it is 0.89 when the  $O_2/CH_4$ 420 mole ratio is 0.8. The higher oxygen carrier circulation at lower oxidation reactor outlet 421 422 temperatures is reflected in the average solids axial velocities being higher.



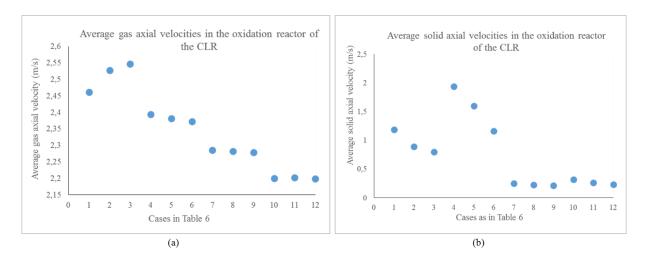




Figure 8: (a) Average gas axial velocity along the bed of the oxidation reactor of the CLR (b) Average solid
 axial velocity along the bed of the oxidation reactor of the CLR

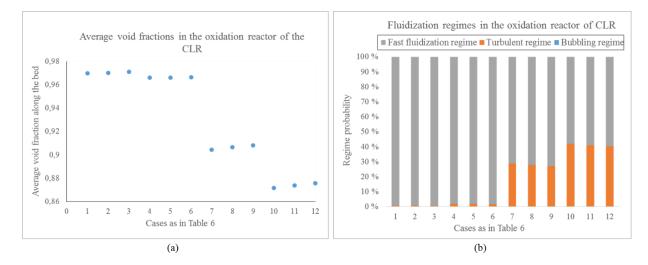


Figure 9: (a) Average void fraction in the oxidation reactor of the CLR (b) Fluidization regime probabilities in the oxidation reactor of the CLR

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# 432 4.2. Technical performance analysis for the CLR-CC process

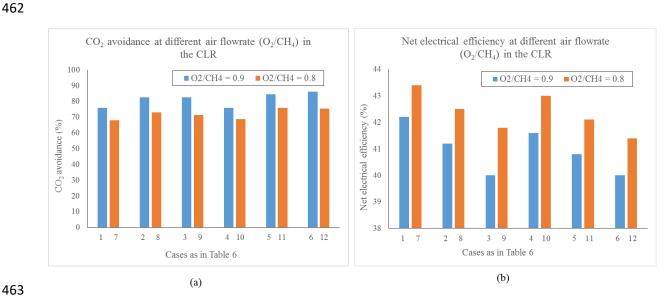
Figure 10 shows the CO<sub>2</sub> avoidance and net electrical efficiency for the CLR-CC process when 433 the O<sub>2</sub>/CH<sub>4</sub> mole ratio is 0.8 and 0.9 in the CLR. The O<sub>2</sub>/CH<sub>4</sub> mole ratio is controlled by varying 434 435 the air flowrate in the oxidation reactor of the CLR. The assumptions in the cases, for which 436 the results are shown in Figure 10, have been defined in Table 6. The CO<sub>2</sub> avoidance in the CLR-CC process is higher by 8-11 % when the  $O_2/CH_4$  is 0.9 in contrast to 0.8 in the CLR. The 437 conversion of CH<sub>4</sub> in the fuel reactor is high when the O<sub>2</sub>/CH<sub>4</sub> is 0.9 resulting in a higher 438 concentration of CO<sub>2</sub> after the WGS step. This helps in producing a H<sub>2</sub>-rich fuel with a higher 439 H<sub>2</sub> purity and lesser concentration of CO and CH<sub>4</sub> (as shown in Table 10) and hence resulting 440 in higher CO<sub>2</sub> avoidance for the CLR-CC process. 441

442 The net electrical efficiency for the CLR-CC process is observed to be higher by ~1.5% -points for the cases with O<sub>2</sub>/CH<sub>4</sub> in the CLR as 0.8 when compared to the cases with O<sub>2</sub>/CH<sub>4</sub> as 0.9 443 (shown in Figure 10). Four components in the CLR-CC process are mainly affected by changing 444 445 the  $O_2/CH_4$  mole ratio in the CLR. When the  $O_2/CH_4$  mole ratio is high (=0.9), the air flowrate to the oxidation reactor is high and hence more work is consumed by the air compressor. Higher 446 air flowrate also implicates higher N<sub>2</sub>-rich stream flow and hence a higher power output from 447 448 the N<sub>2</sub>-rich stream turbine. The GT anyhow gives lesser power output when the O<sub>2</sub>/CH<sub>4</sub> mole ratio is 0.9 when compared to 0.8. It is mainly because the H<sub>2</sub>-rich fuel has a lower composition 449 of CO and CH<sub>4</sub> when the O<sub>2</sub>/CH<sub>4</sub> mole ratio is 0.9 as shown in Table 10. CO (~283 kJ/mol) and 450 CH<sub>4</sub> (~802 kJ/mol) have a higher LHV than the H<sub>2</sub> (~244 kJ/mol). Hence, lower mole 451 composition of CO and CH<sub>4</sub> in the H<sub>2</sub>-rich fuel reflects in lower specific LHV at the inlet of the 452 GT combustion chamber resulting in lower specific power output from the GT. Therefore, the 453 amount of CH<sub>4</sub> at the inlet of fuel reactor of the CLR is high (~170 TPH) in cases with O<sub>2</sub>/CH<sub>4</sub> 454 as 0.9 when compared to 160 TPH of CH4 in the fuel reactor of the CLR in cases with O2/CH4 455 as 0.8 for the process layout of the CLR-CC considered in this paper. Since the mass flowrate 456 of the N<sub>2</sub>-rich stream used as a diluent is same in all the cases, the specific power consumption 457 in the diluent N<sub>2</sub>-rich stream compressor is high when the  $O_2/CH_4$  mole ratio is 0.9. The overall 458

efficiency penalty in the CLR-CC process is therefore high when the O<sub>2</sub>/CH<sub>4</sub> mole ratio in the 459

CLR is 0.9 and hence resulting in a lower net electrical efficiency when compared to cases that 460

have O<sub>2</sub>/CH<sub>4</sub> mole ratio of 0.8 in the CLR. 461



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Figure 10: (a) CO<sub>2</sub> avoidance in CLR-CC at different air flowrates (O<sub>2</sub>/CH<sub>4</sub>) in the CLR (b) Net electrical efficiency of CLR-CC at different air flowrates  $(O_2/CH_4)$  in the CLR

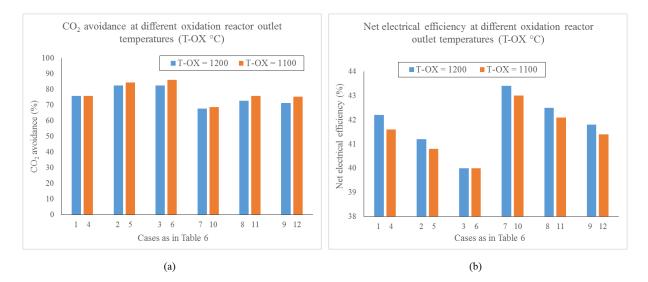
Cases	O <sub>2</sub> /CH <sub>4</sub> by moles	Steam/CH <sub>4</sub> by mass	Oxidation Reactor Outlet Temperature (°C)	H <sub>2</sub> -rich fuel composition (mol%)				
				H <sub>2</sub> O	CO <sub>2</sub>	$CH_4$	СО	$H_2$
1	0.9	0.5	1200	0.5	1.9	0.5	4.5	92.6
2	0.9	1.0	1200	0.5	2.1	1.6	1.4	94.4
3	0.9	1.5	1200	0.5	2.2	2.2	0.6	94.4
4	0.9	0.5	1100	0.5	1.9	0.5	4.5	92.6
5	0.9	1.0	1100	0.5	2.1	0.5	1.7	95.1
6	0.9	1.5	1100	0.5	2.0	1.2	0.7	95.6
7	0.8	0.5	1200	0.5	1.7	4.1	4.2	89.4
8	0.8	1.0	1200	0.5	1.8	5.6	1.2	90.8
9	0.8	1.5	1200	0.5	2.0	7.0	0.5	90.0
10	0.8	0.5	1100	0.5	1.7	2.6	5.1	90.0
11	0.8	1.0	1100	0.5	2.0	3.9	1.5	92.1
12	0.8	1.5	1100	0.5	2.0	5.1	0.6	91.7

466 Table 10: Composition of the  $H_2$ -rich fuel at the inlet of the combustion chamber in the gas turbine system

Figure 11 shows the CO<sub>2</sub> avoidance and net electrical efficiency for the CLR-CC process for 467 different oxidation reactor outlet temperatures. The oxidation reactor outlet temperatures is 468 controlled by the oxygen carrier flowrate in the oxidation reactor of the CLR which does not 469 take part in the oxidation reactions and is only used to transfer heat from the oxidation reactor 470 to the fuel reactor. In this paper, the CLR-CC process was analysed at oxidation reactor outlet 471 temperatures of 1100 and 1200 °C. As seen in Figure 11, the CO<sub>2</sub> avoidance and the net 472 electrical efficiency of the CLR-CC process is less sensitive to the oxidation reactor outlet 473 temperatures. The CO<sub>2</sub> avoidance is higher by 2-4 % in the cases where the oxidation reactor 474 475 outlet temperatures is assumed to be 1100 °C. More heat from the oxidation reactor is transferred to the fuel reactor of the CLR when the oxidation reactor outlet temperature is 1100 476

<sup>477</sup> °C, which results in achieving higher conversion of CH<sub>4</sub> and hence a higher concentration of <sup>478</sup> CO<sub>2</sub> at the absorber inlet of the CO<sub>2</sub> capture section and higher capture and avoidance rates. <sup>479</sup> The net electrical efficiency of the CLR-CC is nearly 0.5% higher when the oxidation reactor <sup>480</sup> outlet temperature is 1200 °C. The difference in net electrical efficiency comes from only one <sup>481</sup> component, the N<sub>2</sub>-rich stream turbine. The temperature of the N<sub>2</sub>-rich stream from the <sup>482</sup> oxidation reactor is same as the oxidation reactor outlet temperature, and hence higher the <sup>483</sup> temperature, higher is the power output from the turbine.







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Figure 11: (a) CO<sub>2</sub> avoidance in CLR-CC for different oxidation reactor outlet temperatures (b) Net electrical efficiency of CLR-CC for different oxidation reactor outlet temperatures

488 Figure 12 shows the CO<sub>2</sub> avoidance and net electrical efficiency of the CLR-CC process for 489 different steam/CH<sub>4</sub> ratio by mass in the fuel reactor of the CLR. The CO<sub>2</sub> avoidance for the 490 CLR-CC process is 4-11 % higher when the steam/CH<sub>4</sub> ratio (by mass) is varied between 0.5, 1 and 1.5 in the fuel reactor of the CLR. Availability of the steam in the fuel reactor not only 491 restricts the coke formation on the oxygen carrier in the fuel reactor, but also enhances the 492 493 conversion of CO into CO<sub>2</sub> through a WGS reaction. Hence, the concentration of CO<sub>2</sub> is high in the syngas (as seen in Table 9) and the stream at the absorber inlet of the CO<sub>2</sub> capture section, 494 when the steam/CH<sub>4</sub> ratio in the fuel reactor is high, resulting in a higher CO<sub>2</sub> avoidance rate 495 for the CLR-CC. The net electrical efficiency of the CLR-CC decreases by ~1% for every 0.5 496 point increase in the steam/CH<sub>4</sub> mass ratio in the fuel reactor. The ST in the power plant directly 497 affects the net electrical efficiency. Steam for the fuel reactor is extracted from the MP steam 498 turbine. Therefore, at higher steam/CH<sub>4</sub> mass ratio in the fuel reactor, more amount of steam is 499 extracted from the ST resulting in lower power output from the ST system. 500

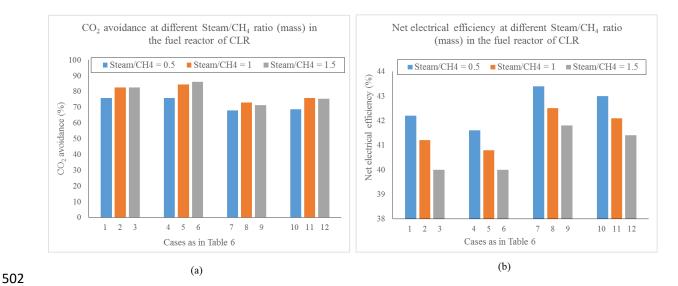


Figure 12: (a) CO<sub>2</sub> avoidance in CLR-CC at different steam/CH<sub>4</sub> mass ratio in the fuel reactor of CLR (b) Net
 electrical efficiency of CLR-CC at different steam/CH<sub>4</sub> mass ratio in the fuel reactor of CLR

## 506 4.3.Economic analysis of the CLR-CC process

The main results from the economic analysis for the CLR-CC are shown in Table 8 for different 507 cases studied in this paper. Figure 13 shows the contribution of TCR, FOM, VOM and FC to 508 the LCOE of the CLR-CC whereas Figure 14 shows the contribution of capital costs of different 509 sections in the process to the BEC. The LCOE for the CLR-CC process is 4-7 \$/MWh less when 510 the O<sub>2</sub>/CH<sub>4</sub> mole ratio in the CLR is 0.8 when compared to the cases having O<sub>2</sub>/CH<sub>4</sub> mole ratio 511 of 0.9 in the CLR. The main difference is observed in the TCR and the FC. For O<sub>2</sub>/CH<sub>4</sub> mole 512 ratio of 0.9 in the CLR, the flowrate and temperature of syngas is high. Hence, the amount of 513 steam produced in cooling of syngas and the product streams from HTS and LTS is high, which 514 demands more heat exchange area. Therefore, as seen in Figure 14Error! Reference source 515 not found., the BEC of the components associated with N<sub>2</sub>-rich stream treatment is higher. 516 Although less significant, but the VOM is slightly more when the O<sub>2</sub>/CH<sub>4</sub> mole ratio is 0.9 in 517 the CLR. This is mainly due to the costs incurred by higher requirement of oxygen carrier. The 518 CLR-CC process also requires higher CH<sub>4</sub> input to the fuel reactor for the cases with O<sub>2</sub>/CH<sub>4</sub> 519 mole ratio of 0.9, and hence the FC is high when the  $O_2/CH_4$  mole ratio is 0.9. 520

The LCOE of the CLR-CC is 3-6 \$/MWh less for the cases with oxidation reactor outlet temperature of 1200 °C when compared to the cases for which the temperature is 1100 °C. The difference in LCOE is due to the higher VOM in the cases with oxidation reactor outlet temperature of 1100 °C because of the higher oxygen carrier utilization in the CLR. There is no significant difference in the other components of the LCOE at different oxidation reactor outlet temperatures.

527 The steam/CH<sub>4</sub> mass ratio in the CLR has less effect on the LCOE of the CLR-CC process. The 528 LCOE of the CLR-CC changes by  $\sim$ 1 \$/MWh for a change of 0.5 in the steam/CH<sub>4</sub> mass ratio 529 in the fuel reactor of the CLR. The LCOE is less for the cases with higher net electrical 530 efficiency because the fuel consumption for 1MWh electricity produced is less, and hence the 531 FC component of the LCOE is lower.

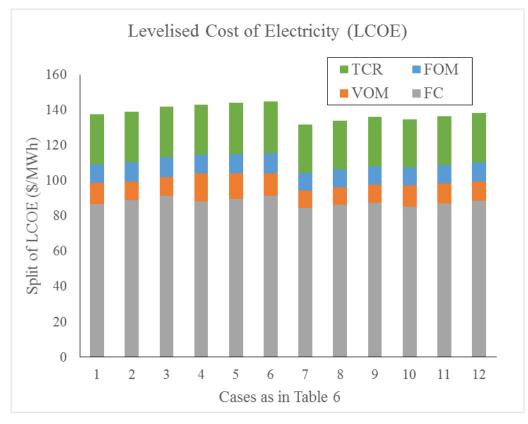


Figure 13: Contribution of the TCR, FOM, VOM and FC to the LCOE of the CLR-CC process for different cases
 defined in Table 6

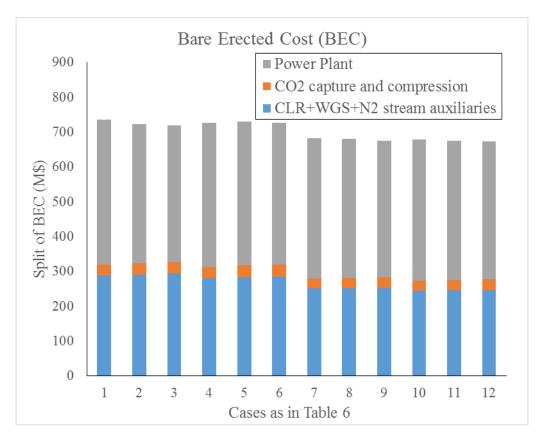




Figure 14: Bare erected cost for the CLR-CC for different cases defined in Table 6

540 4.4. Effect of fuel cost and process contingency on LCOE

541 Error! Reference source not found. Figure 13 clearly shows that the major contributors to the LCOE of the process is the fuel costs and the TCR. While estimating the TCR of the CLR-CC 542 process, the process contingency was assumed 50% of BEC, which is for a process which is 543 considered to be a new concept with limited data (GCCSI 2013). The fuel cost was assumed 544 10.18 \$/GJ-LHV. Anyhow, the process contingency of the process depends completely on its 545 level of maturity whereas the fuel cost is very much dependent on the region from where it is 546 imported. Error! Reference source not found. Figure 15 provides a sensitivity study for the 547 defined cases when the process contingency is 50% and 10% of BEC, and the fuel cost is 10.18 548 and 4.5 \$/GJ-LHV. As seen in Figure 15Error! Reference source not found., the LCOE for 549 the CLR-CC process varies between 75.3 and 144.8 \$/MWh. 550



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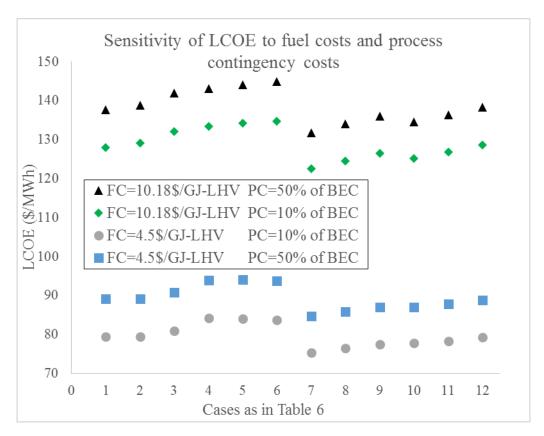






Figure 15: Sensitivity of LCOE to fuel cost and process contingency

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# 555 **5.** Conclusion

This paper presents the techno-economic analysis of the CLR-CC process. The conditions in the CLR were simulated using the 1-D phenomenological model developed by Francisco Morgado et al. (2016). The process layout for the CLR-CC was discussed and a sensitivity study with respect to the pressure inside the CLR was presented by Nazir, Bolland, and Amini (2018). In this paper, three manipulative variables, air flowrate (O<sub>2</sub>/CH<sub>4</sub> molar ratio) at the inlet of oxidation reactor, oxidation reactor outlet temperature and steam flow rate (steam/CH<sub>4</sub> mass

ratio) to the fuel reactor, were selected. The effect of changes in these variables on the overall 562 techno-economic performance of the CLR-CC process was analysed over 12 different cases. 563 The main techno-economic performance indicators of the process are CH<sub>4</sub> conversion in the 564 fuel reactor, CO<sub>2</sub> avoidance rates, net electrical efficiency and the LCOE. The CH<sub>4</sub> conversion 565 in the fuel reactor is between 85-99% and is highly sensitive to the air flowrate ( $O_2/CH_4$  mole 566 ratio) in the CLR. The CO<sub>2</sub> avoidance rates in the CLR-CC is between 67-86%. The CO<sub>2</sub> 567 avoidance is highly sensitive to the air flowrate (O<sub>2</sub>/CH<sub>4</sub> mole ratio) in the oxidation reactor 568 followed by the steam flow rate (steam/CH<sub>4</sub> mass ratio) in the fuel reactor of the CLR. The net 569 electrical efficiency of the CLR-CC is between 40-43.5% for the cases studied in this paper. 570 The net electrical efficiency is sensitive to the O<sub>2</sub>/CH<sub>4</sub> mole ratio in the CLR and the steam/CH<sub>4</sub> 571 mass ratio in the fuel reactor of the CLR. The net electrical efficiency is higher when the O<sub>2</sub>/CH<sub>4</sub> 572 mole ratio in the CLR is 0.8 when compared to 0.9, and the net electrical efficiency decreases 573 574 with an increase in steam/CH<sub>4</sub> mass ratio in the fuel reactor. The LCOE for the CLR-CC lies between 131-145 \$/MWh. The LCOE of the CLR-CC is less sensitive to the process parameters 575 and is highly sensitive to the NG price (fuel costs) followed by the process contingencies cost. 576 The LCOE is reduced by 40% if the NG price is halved whereas if the process contingency is 577 reduced from 50% (for a new technology) to 10% (more mature and commercially available 578 technology), the LCOE reduces by  $\sim 10\%$ . 579

The CO<sub>2</sub> avoidance in the CLR-CC is on par with other pre- and post-combustion capture 580 methods. Anyhow, the net electrical efficiency of the CLR-CC for the conditions reported in 581 this paper is close to the net electrical efficiency of a combined cycle with steam-methane 582 reforming (~43.6%) but is less than that of combined cycles with auto-thermal reforming 583 (~46.9) and post-combustion capture methods involving chemical absorption (~49-50%). The 584 efficiency of the CLR-CC process is also limited by the gas turbine design. One of the major 585 benefits from the CLR-CC process when compared to the post-combustion capture methods is 586 the flexibility in operating the plant based on the needed output which could be H<sub>2</sub> or electric 587 power. The current analysis helps in understanding the trends and techno-economic behavior 588 of the CLR-CC process for the chosen manipulated variables. Improving efficiency of the 589 process will help in reducing fuel consumption and hence reducing the costs. Therefore, further 590 optimization studies for the CLR-CC process is suggested. The developed methods in this paper 591 can also be applied to analyzing novel reactor concepts for reforming like the gas-switching 592 reforming (Wassie et al. 2017). 593

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# 596 6. Acknowledgements

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