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Full Plant Scale Analysis of Natural Gas Fired Power Plants with Pre-Combustion CO_2 Capture and Chemical Looping Reforming (CLR)

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Abstract

In this study, first of its kind complete plant scale integration of pre-combustion CO_2 capture method with Chemical Looping Reforming (CLR) of Natural Gas (NG), Water Gas Shift (WGS) process, CO_2 capture and CO_2 compression in a combined cycle power plant has been presented. The CLR consisted of oxidation and fuel reactor. The oxidation reactor oxidizes the metal oxygen carrier with compressed air and produces an oxygen depleted air stream (N_2 stream) as by-product. The fuel reactor reforms the NG with the metal oxide in presence of steam to produce syngas. The syngas is further subjected to WGS and CO_2 capture using a-MDEA, to prepare a H_2-rich fuel, which is combusted in the Gas Turbine (GT) system. The heat from cooling of process streams in the pre-combustion CO_2 capture method, is used to prepare saturated low pressure steam, fraction of which is used in reboiler to regenerate the amine for CO_2 capture, and the remainder is expanded in Steam Turbine (ST) to generate power. The power plant is a combined cycle with two GT, two Heat Recovery Steam Generators (HRSG) and one ST. 12% of air entering the GT is used in the oxidation reactor of CLR, and equivalent amount of N_2 stream is compressed and added as diluent in the GT. The overall process was integrated and analysed at full load conditions. The current process has also been compared with Natural Gas Combined Cycle (NGCC) plant without CO_2 capture. The net electric efficiency of the power plant with pre-combustion CO_2 capture in this study is 43.1%, which is 15.3%-points less than the NGCC plant without capture. Major energy penalty in the process comes from air compressor, the diluent N_2 stream compressor and due to low degree of process integration to avoid complexity.

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1. Introduction

The need for CCS in large one-point source emissions like power plants has been well debated and established. CCS is foreseen to contribute with one sixth of total CO_2 emission reductions required by the year 2050, as stated by International Energy Agency (IEA) in the "Energy Technology Perspectives 2012". The awareness has resulted in growing scientific research in post-, pre- and oxy-combustion capture routes for CO_2 capture. Boot-Handford, Abanades [1] gave an update of the developments in these capture routes. Although post-combustion amine absorption is the most mature technology, chemical looping processes possess an attractive thermodynamic potential.

Chemical-looping processes (Chemical Looping Combustion (CLC) and Chemical Looping Reforming (CLR)) with its ability to inherently separate CO_2 has gained significant research attention. The concept was first proposed by Richter and Knoche [2]. The first of its kind CLC based power generation cycle was proposed by Ishida et al. [3, 4]. CLC effectively converts the chemical energy of fossil fuel into heat at a fairly low temperature (T ≈ 800-900 °C) [5-7], which limits the efficiency of the power generation process. Compared to conventional combustion processes, On the other hand, CLR converts the chemical energy of fossil fuel into chemical energy of a H_2-rich fuel, which results in streams with high temperature (T ≈ 1400-1500 °C) after combustion in a gas turbine system.

Figure 1 shows the block diagram of a typical CLR process. Natural gas (NG) undergoes partial oxidation upon reaction with metal oxide in a fuel reactor to produce syngas, which is mainly a mixture of CO, CO_2, H_2 and H_2O. Steam is added in the fuel reactor to maintain a sufficient steam to carbon ratio to avoid coke formation and also to provide favorable conditions for downstream water gas shift reactions. The reduced metal oxygen carrier is oxidized with air in the oxidation reactor, which also results in an oxygen depleted air stream (N_2 stream).

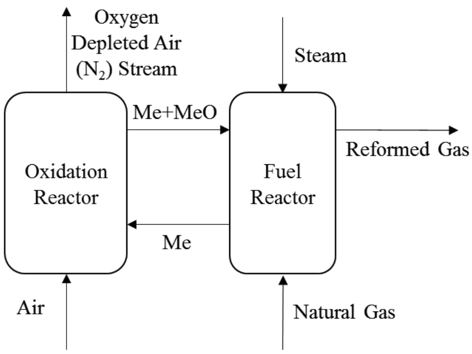

Fig. 1. Flow diagram for a typical chemical looping reforming process

Numerous studies have been carried out on the choice of oxygen carrier [8, 9], reactor scale modeling and experimental studies [10-14]. Studies on power generation processes with Ca-Cu looping [15], auto-thermal reforming [16, 17] have also been reported. Cormos, Petrescu [18] studied the performance of chemical looping systems with CO_2 capture with more focus on hydrogen production. Kvamsdal, Jordal [19] studied the performance of different CO_2 capture processes integrated with power plants, including the pre-combustion capture routes with auto-thermal reforming and hydrogen separating membrane reactor.

There is still a gap in knowledge with respect to information on behavior of the power plant by integrating it with CLR and CO_2 capture. The objective of this paper is to fill this gap by presenting the first of its kind complete plant scale integration of pre-combustion CO_2 capture method with chemical looping reforming of NG, WGS process, CO_2 capture and CO_2 compression in a combined cycle power plant. Mass and energy balances for the process are established to evaluate the performance of the power generation process. Net electrical efficiency is chosen as a key performance indicator. The current paper also discusses the energy penalty in the process. The results have been compared with a Natural Gas Combined Cycle (NGCC) plant without CO_2 capture. Section 2 of the paper describes the process. Section 3 describes the methodology and the assumptions used in the study. The results and discussion are presented in Section 4 and conclusions in Section 5.

2. Process Description

The current study is about a pre-combustion CO_2 capture route in natural gas fired power plants, where natural gas is reformed and converted to H_2-rich fuel, which is then subjected to CO_2 capture before being combusted in a gas turbine system. The power generation cycle is similar to a the cycle proposed in EBTF [20] for a NGCC plant without CO_2 capture and consists of two Gas Turbines (GT) with two Heat Recovery Steam Generators (HRSG) and one triple pressure Steam Turbine (ST) and a condenser. One natural draft cooling tower is used to supply cooling water to the entire process. The pre-combustion capture route consists of a CLR comprising of interconnected oxidation and fuel reactor, High and Low Temperature (HT and LT) Water Gas Shift reactors (WGS), CO_2 separation using amine absorption and CO_2 compression stages.

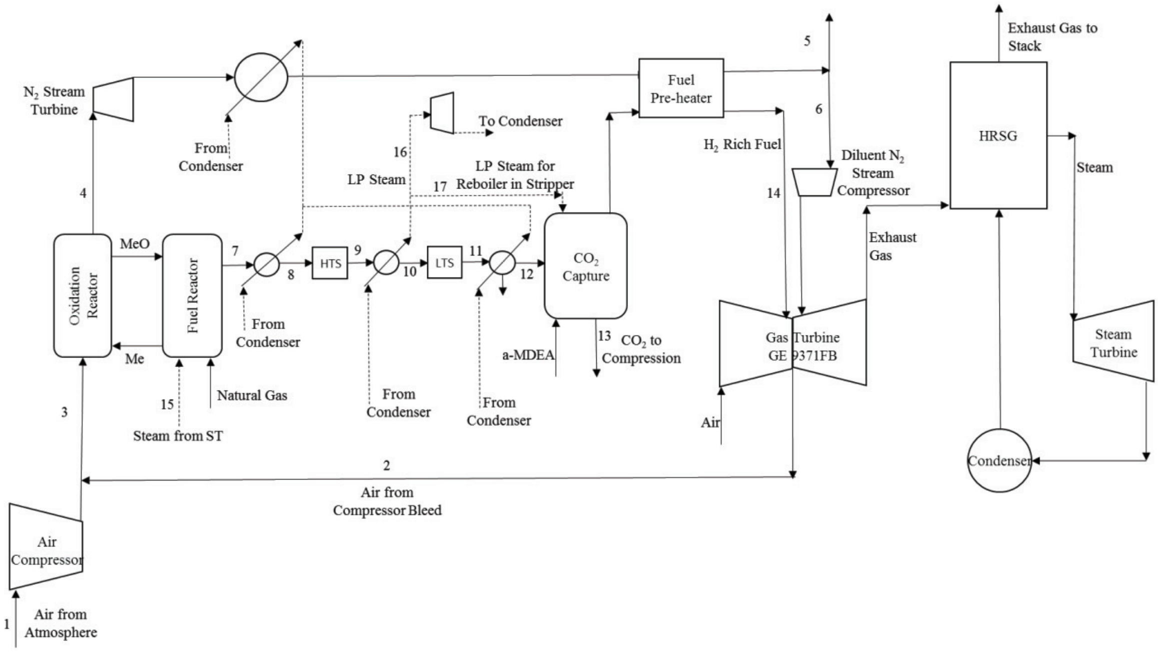

Fig. 2. Process layout for pre-combustion CO_2 capture using CLR

Figure 2 shows the process flow diagram of the proposed process. The CLR is operating at 18 bar with a pressure drop of 5% in both oxidation and fuel reactor. This reactor pressure is very close to the discharge pressure of compressor air bleed from the GT system. Therefore, the compressor bleed need not be expanded or compressed before being used in the CLR. Natural gas is partially oxidized with metal oxide (FeO) in the fuel reactor of the CLR, as given in reaction (1). Steam is added to the fuel reactor to minimize the possibility of coke formation and maintain favorable conditions for the proceeding WGS steps. Steam used in the fuel reactor is extracted from the MP steam turbine. The reduced oxygen carrier is sent to the oxidation reactor and reacted with compressed air (reaction (2)). Oxygen depleted air stream (N_2 stream) is the by-product from the oxidation reactor. The N_2 stream is expanded in a gas/air turbine to near atmospheric pressure before being cooled down to produce LP steam. A major fraction of the

N_2 stream (equal to the flowrate of compressor air bleed from GT) is sent to the GT system as diluent and the remainder is vented out.

$$C_xH_y + x\ MeO \rightarrow x\ CO + \frac{y}{2}\ H_2 + x\ Me \qquad \Delta H > 0\ kJ/mol \qquad (1)$$

$$x\ Me + \frac{x}{2}\ O_2 \rightarrow x\ MeO \qquad \Delta H < 0\ kJ/mol \qquad (2)$$

$$CO + H_2O \leftrightarrow CO_2 + H_2 \qquad \Delta H = -41\ kJ/mol \qquad (3)$$

The syngas from the fuel reactor is cooled down and passed on to high and low temperature water gas shift reactors in series. CO and H_2O is converted to CO_2 and H_2 through a water gas shift reaction (3), which is exothermic. The water gas shift reaction is driven catalytically in two stages to get higher conversion at lower steam requirements [21]. The resulting stream from the WGS section is cooled and subjected to CO_2 capture through chemical absorption using piperazine activated 45 wt% Methyl diethanolamine (a-MDEA). CO_2 is absorbed into the solvent in the absorber section and the solvent is regenerated in the stripper, where LP steam is used in the reboiler for regeneration. The MDEA absorption technique serves well at moderate partial pressures of CO_2 which is the case in this study [22]. The choice of CO_2 capture method is not in the scope of the current study. The captured CO_2 is then compressed to 110 bar and is ready for transport and storage.

The H_2-rich fuel after CO_2 separation is compressed and combusted with air in the GE 9371FB gas turbine system [20]. N_2 stream from the oxidation reactor is used for dilution which not only compensates for the mass of compressor bleed but also helps in reducing the peak flame temperatures and NO_x formation when hydrogen is combusted [23]. The gas turbine exhaust passes through the HRSG to produce steam for the steam cycle before being released into the atmosphere. The steam cycle is a three-pressure level with reheat at 166/37/3.8 bar for HP/MP/LP type of steam, respectively. The water and steam mixture from the ST system is condensed in a condenser to prepare feed water for the steam cycle. Natural draft cooling tower supplies the cooling water to the condenser.

The heat from N_2 stream, syngas and WGS reaction is used to prepare saturated LP steam. The reboiler of the stripper uses saturated LP steam to regenerate the amine. The remainder of the LP steam is used to produce work in a separate steam turbine (additional LP steam turbine).

3. Methodology

Net electric efficiency (η) on Lower Heating Value (LHV) basis of the fuel, is chosen as the parameter that defines the performance of the power plant. It is defined as:

$$\eta = \frac{\text{Net electricity produced}}{\text{LHV of NG input to the process}}$$

The assumptions and methodology followed is shown for the three sections of the process (a) CLR (b) WGS, CO_2 capture and compression (c) power plant.

3.1. CLR

The mass and heat balance calculations at equilibrium for air compressor and CLR were carried out in Aspen Plus V8.6, since it contains the property set for solids taking part in the reactions in CLR [14]. The Peng Robinson model was used to estimate the equilibrium conditions in the compressor and CLR at steady state [14]. The work in compressing air was estimated using the ASME Method in Aspen Plus, which uses polytropic efficiency. The Gibbs Reactor Module in Aspen Plus was used to simulate the adiabatic conditions in oxidation and fuel reactor. The stream class was specified as MIXCISLD to accommodate for the presence of conventional solid metal particles without specifying the particle size distribution.

The compositions and conditions for air and natural gas has been taken from the EBTF [20] and is shown in Table 1 and 2. Atmospheric air is considered to be at 15 °C and 1.01325 bar. Natural gas is delivered at 10 °C and 70 MPa.

Table 1. Composition of Air [20].

Component	Volume Fraction dry	Volume Fraction at 60% relative humidity
N_2	78.09	77.30
CO_2	0.03	0.03
H_2O	1.01	0
Ar	0.932	0.923
O_2	20.95	20.74
Gas Constant (J/kg K)	287.06	288.16
Molecular Weight	28.964	28.854

Table 2. Composition of Natural Gas [20]

Component	Volume %
CH_4 – Methane	89
C_2H_6-Ethane	7
C_3H_8 – Propane	1
C_4 – i-Butane	0.05
C_4 – n-Butane	0.05
C_5 – i-Pentane	0.005
C_5 – n-Pentane	0.004
CO_2	2
N_2	0.89
S	<5 ppm
HHV (MJ/kg)	51.473
LHV (MJ/kg)	46.502

Atmospheric air is compressed to 18 bar in a compressor, for which 92% polytropic efficiency and 95% mechanical efficiency is assumed. This compressed air is mixed with air bled from the GT. 12% of the air entering the GT system is extracted as compressor bleed and used in oxidation reactor. The amount of total air entering the oxidation reactor is set based on the required conversion of NG in fuel reactor. FeO has been considered as the oxygen carrier in this study since it is inexpensive, has high melting point and non-toxic [8]. It is assumed that all the oxygen entering the oxidation reactor, reacts with FeO and forms Fe_3O_4. The FeO flow rate entering the oxidation reactor is set to 68 tons/ton of NG. An excess of metal flow rate is considered to keep the overall temperature in the oxidation reactor below the melting point of the oxygen carrier and close to 1200 °C. A pressure drop of 5% is assumed in both the oxidation and fuel reactor. After the reaction, the N_2 stream is sent to the gas/air turbine to produce power and the FeO-Fe_3O_4 mixture is sent to the fuel reactor.

NG is reacted with Fe_3O_4 to produce syngas in the fuel reactor at adiabatic conditions. Steam at 18 bar and 283 °C is added to the fuel reactor with the flow rate of 1 ton/ton of NG, and hence a steam to carbon ratio of 1.38 is maintained at the inlet of fuel reactor. 99% conversion of CH_4 in NG is assumed. Based on these conditions, flow rate of air entering the oxidation reactor is set, which is 6.4 tons/ton of NG. The syngas is then sent to the WGS and the reduced metal oxide is circulated back to the oxidation reactor.

3.2. WGS, CO_2 Capture and Compression

The WGS, CO_2 Capture and Compression processes were modeled in Aspen HYSYS V8.6. The equilibrium reactor module was selected to simulate the catalytic conditions for WGS at high and low temperature. The two-step WGS was simulated adiabatically with feed streams at 400 °C and 200 °C. The pressure drop in both the WGS steps was assumed to be 0.5 bar. The main reaction in the WGS steps is reaction (3). The heat exchangers in the process are assumed to have a pressure drop of 0.4 bar if the fluid is liquid, and 2% if the fluid is a gas [20]. Saturated LP steam at 3.8 bar is produced while cooling the HT and LT WGS product streams. This steam is used in a ST to produce power.

The absorber and regenerator conditions are given in Table 3. 95% CO_2 capture rate in the absorber is assumed and accordingly the flow rate of amine is set. The optimization of the CO_2 capture process is not in the scope of this study and hence the main results from this section are the H_2 rich fuel quality and reboiler duty in the stripper. The reboiler duty of the stripper obtained is 1.48 MJ per kg CO_2 captured in the process. This is very close to the assumption made by Nord, Anantharaman [16] for the absorption process using MDEA in pre-combustion capture. Saturated steam at 3.8 bar is used in the reboiler. The captured CO_2 is then compressed and pumped to 110 bar before making it ready for transport and storage. The compression steps have been simulated as suggested in EBTF [20].

Table 3. Absorber and regenerator conditions.

Number of absorber trays	20
Number of stripper trays	20
Pressure drop in the absorber (bar)	0.1
Pressure drop in the stripper (bar)	0.1
Lean amine loading (mol CO_2/mol MDEA)	0.301
Rich Amine Loading (mol CO_2/mol MDEA)	0.666
Lean Amine Flowrate (Std Liq Flow) (m3/s)	1.55
Condenser Temperature in Stripper (°C)	46.11
Reboiler Duty (MJ/kg CO_2 separated in stripper)	1.95
Reboiler Duty (MJ/kg CO_2 captured)	1.48

3.3. Power Plant

The combined cycle power plant has been modeled using the Thermoflex component of Thermoflow Suite [24]. Thermoflow holds a database of commercial gas turbines, but GE 9371FB has been chosen for the power plant in the current study. The chosen GT is robust to changes in fuel type and also supports fuel which is rich in hydrogen [16, 20]. The power plant comprises of two GT and HRSG connected to a single ST system. The steam cycle is with three pressure levels at 166/37/3.8 bar. The GT is run at 100% load. Based on these conditions, the amount of fuel input is estimated. This determines the flow rate of syngas entering the WGS and the flow rate of NG entering the fuel reactor in the CLR. 12% of the air entering the GT system is extracted as compressor bleed and used in the oxidation reactor of the CLR.

4. Results and Discussion

The main results of the full plant scale analysis is summarized in Table 4 and the stream data in Table 5. Table 4 includes the comparison of two different power generation processes: (i) Power Plant with CLR, WGS, CO_2 capture and compression (PP with CLR & CO_2 Capture) (ii) NGCC without CO_2 capture. Gross power outputs from different generator terminals, the net power output, the power consumption in the fuel, air and N_2 stream compressors and in auxiliaries, and net electrical efficiencies are listed in the table. The auxiliaries include the GT and ST auxiliaries and boiler feed water (BFW) pumps. In the process with CLR and CO_2 capture, additional auxiliaries include the pumps in heat recovery from reforming and water gas shift steps.

The net plant efficiency for the power plant with CLR and CO_2 capture and compression at 100% load is 43.1% with a net electrical output of 877 MW. The net power output is estimated by subtracting the power consumed in air compressor, fuel compressor, N_2 stream compressor, CO_2 compression, pump for regenerated amine, auxiliaries from gross power output from the generator terminals. The current process under study is compared to the reference NGCC plant without CO_2 capture, with the design conditions specified in EBTF [20].

Table 4. Comparison of results for power plant with CLR and CO_2 capture and NGCC without CO_2 Capture.

		PP with CLR & CO_2 Capture	NGCC without CO_2 Capture
Turbine Inlet Temperature	°C	1433	1427
Turbine Exhaust Temperature	°C	642	644
Gas Turbine	MW	607.4	579.2
Steam Turbine	MW	291.8	334.9
N_2 Stream Turbine	MW	173.4	-
Work from expansion of extracted MP steam for reforming	MW	6.9	-
Additional LP steam Turbine	MW	53.3	-
Diluent N_2 Stream Compressor	MW	108.7	-
Diluent N_2 Stream Compressor - % of LHV Input	%	5.34	-
H_2 rich fuel Compressor	MW	27	-
H_2 rich fuel Compressor - % of LHV Input	%	1.33	-
Air Compressor	MW	55.2	-
Air Compressor - % of LHV Input	%	2.71	-
Pump for Regenerated Amine	MW	2.6	-
Pump for Regenerated Amine - % LHV Input	%	0.13	-
CO_2 Compressors and Pump	MW	37.5	-
CO_2 Compressors and Pump - % LHV Input	%	1.84	-
Energy (kWhr) to compress CO_2 captured	kWhr/kg CO_2	0.1	
Auxiliaries	MW	24.7	19.5
Auxiliaries - % LHV Input	%	1.21	1.29
Net Electrical Output	MW	877	883
Mass of NG Input	TPH	157.4	117.1
LHV of NG – Input	MW	2034	1513
Net Electrical Efficiency	%	43.1	58.4
CO_2 Avoidance	%	84.5	-
CO_2 Capture	%	88.5	-

The net electrical efficiency of the reference NGCC plant without CO_2 capture is 58.4% with net electrical output 883 MW. The power plant with CLR and CO_2 capture and compression experiences efficiency penalty of 15.3%-points compared to the reference plant without capture. A number of factors and considerations cause the large efficiency penalty in the power plant with CLR and CO_2 capture. The compressors and pumps in the CO_2 capture and compression section account for a 1.97%-points of energy penalty from the net NG LHV input to the process. The compressor to pressurize the H_2 rich fuel and the N_2 stream to the conditions in the GT system accounts for energy penalty of 1.33% and 5.34%-points respectively. The air compressor to supply additional air to the oxidation reactor accounts for 2.71%-points of energy penalty. In other pre-combustion processes reported in literature, for example, ATR-IRCC cycle [16, 17], compressed air bleed from the GT is sufficient for reforming. There is a balance between the power consumed in compressors for diluent N_2 stream, H_2 rich fuel and air with the power generated from the N_2 stream turbine, additional LP steam turbine and expansion of MP steam extracted from the ST system. These compressors and turbines can be mounted on a single shaft to reduce motor and generator losses.

Table 5. Stream data for power generation process with CLR and CO_2 Capture (stream numbers from Fig 2.)

Stream	P (bar)	T (°C)	Flow (TPH)	H_2O mol%	CO_2 mol%	CH_4 mol%	CO mol%	H_2 mol%	N_2 mol%	O_2 mol%	Ar mol%
1	1.01	15	453	1.01	0.03	-	-	-	77.3	20.74	0.92
2	18	416,7	555	1.01	0.03	-	-	-	77.3	20.74	0.92
3	18	416,7	1008	1.01	0.03	-	-	-	77.3	20.74	0.92
4	17.1	1199.3	776	1.22	0.03	-	-	-	97.52	-	1.16
5	1.02	73.8	221	1.22	0.03	-	-	-	97.52	-	1.16
6	1.02	73.8	555	1.22	0.03	-	-	-	97.52	-	1.16
7	16.25	984.5	547	31.09	8.31	0.17	17.87	42.34	0.22	-	-
8	15.60	400	547	31.09	8.31	0.17	17.87	42.34	0.22	-	-
9	15.10	503.7	547	21.54	17.86	0.17	8.33	51.89	0.22	-	-
10	14.79	200	547	21.54	17.86	0.17	8.33	51.89	0.22	-	-
11	14.29	278.4	547	14.85	24.55	0.17	1.64	58.58	0.22	-	-
12	13.73	50	456	0.98	28.54	0.19	1.90	68.13	0.25	-	-
13	110	25	372	0.25	99.42	-	-	0.32	-	-	-
14	13.26	140	82	0.59	1.78	0.27	2.64	94.38	0.35	-	-
15	18	283	157	1	-	-	-	-	-	-	-
16	3.8	141.8	380	1	-	-	-	-	-	-	-
17	3.8	141.8	253	1	-	-	-	-	-	-	-

In the current study, the HRSG design is not modified so as to avoid complexity in process integration. The heat from the N_2 Stream, the reforming and water gas shift reactions is used to produce 633 TPH of saturated LP steam at 3.8 bar, of which 253 TPH is used in the reboiler of the stripper in the CO_2 capture section. The remaining 380 TPH of LP steam is expanded in a steam turbine to produce work. The total rate of heat transfer in cooling syngas, HT and LT WGS product streams and N_2 stream to produce 380 TPH of saturated LP steam is 274 MW. Anyhow, only 53.3 MW power is generated when 380 TPH of saturated LP steam is expanded in a steam turbine. There is a 10.85 %-points energy efficiency loss in the process of converting the heat from process streams into electricity. Better heat integration to produce steam at three different levels and integrating them with the HRSG network will improve the net plant efficiency. Studies related to process improvement with heat integration and new HRSG design conditions is not in the scope of this paper.

NG used in the pre-combustion capture process is 34.4% more than the amount used in reference NGCC power plant without capture. The excess NG in the CO_2 capture route is because of the energy losses at various points in the process apart from maintaining the full load conditions and similar LHV input at the GT inlet. The CO_2 avoidance rate is 84.5% and is defined as the ratio of CO_2 avoided in the process and the CO_2 emitted by the reference plant without CO_2 capture. The CO_2 capture rate of the overall process is 88.5%. The CO_2 capture rate is defined as the fraction of CO_2 formed which is captured and compressed for storage.

5. Conclusions

This paper investigates a natural gas based power plant with pre-combustion capture of CO_2 through CLR, WGS and chemical absorption using a-MDEA. The mass and heat balances for the overall process was established, and the net electrical efficiency was evaluated and compared to a reference NGCC plant without CO_2 capture. The net electrical efficiency estimated for the current process is 43.1%, which is 15.3%-points less than the NGCC plant without capture. Efficiency losses between 8 and 16%-points have been reported in literature for pre-combustion capture processes in natural gas based power plants [16, 17, 25]. The major energy loss in the current process comes from the air compressor (2.71%), diluent N_2 stream compressor (5.34%) and converting the heat from syngas, N_2 stream, HT and LT WGS product streams into electricity.

A fairly low level of heat integration is present in the current process with pre-combustion capture of CO_2, which is reflective through not changing the HRSG design, producing saturated LP steam from cooling of syngas, HT and LT WGS product streams and N_2 stream, and using a separate steam turbine to generate power from saturated LP steam. Although the low degree of integration avoids complexity, it effects the overall efficiency of the process. The energy efficiency loss in producing saturated LP steam and producing power from it is 10.85%-points. Improvements in heat integration and a modified HRSG design will improve the net plant efficiency.

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