NOVEL CONCEPTS FOR NEAR-ZERO EMISSIONS IGCC POWER PLANTS

by

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The paper aims in examining and evaluating the state-of-the-art in techno-logical concepts towards zero-emission coal-fired power plants. The dis-cussion is based on the evaluation of a novel concept dealing with the car-bonation-calcination process of lime for CO_2 capture from coal-fired power plants, compared to the integration of CO_2 capture in an Integrated Gasifi-cation Combined Cycle power plant. Results from thermodynamic simula-tions dealing with the most important features for CO_2 reduction are pre-sented. Preliminary economic considerations are made, taking into account investment and operating costs, in order to assess the electricity cost related to the two different technological approaches.

The cycle calculations were performed with the thermodynamic cycle calcu-lation software ENBIPRO (ENergie-BIllanz-PROgram), a powerful tool for heat and mass balance solving of complex thermodynamic circuits, calcula-tion of efficiency, exergetic and exergoeconomic analysis of power plants. The software code models all pieces of equipment that usually appear in power plant installations and can accurately calculate all thermodynamic properties at each node of the thermodynamic circuit, power consumption of each component, flue gas composition etc. [1]. The code has proven its validity by accurately simulating a large number of power plants and through comparison of the results with other commercial software.

Key words: *IGCC power plants, CO_2 capture*

Introduction

According to the third assessment report of the Intergovernmental Panel on Cli-mate Change (IPCC), most of the observed global warming over the last 50 years is likely to have been due to the increase in greenhouse gas concentrations in the atmosphere. In identifying strategies for mitigation of climate change, it is concluded that combinations of multiple technologies in all sectors must be considered [2]. Among these, CO_2 seques-tration holds an important position. In this context, the integration of CO_2 sequestration concepts in power generation schemes is currently a field of intensive study.

In the frame of CO_2 capture and sequestration in power plants, this paper con-tributes to the investigation of a new pre-combustion concept based on CO_2 removal dur-

ing coal gasification by means of CaO. This technology implies the production of a low-carbon fuel gas in one reactor by integrating all the processes, namely gasification, CO shift and CaO carbonation, in a single step. The proposed process appears to have energy and efficiency penalties lower than the CO_2 pre-combustion capture with physical absorption by means of Selexol.

Technology description

In the frame of CO_2 pre-combustion capture, coal is gasified in order to produce synthesis gas, which is mainly a mixture of H_2 and CO. In the conventional coal gasification process, coal enters the gasifier with steam and oxygen or air, in a high temperature and pressure atmosphere. The products of conventional gasification are syngas and a solid ash waste product [3]. This process is described by the following reaction:

$$2C \quad H_2O \quad \frac{1}{2}O_2 \quad 2CO \quad H_2 \qquad (1)$$
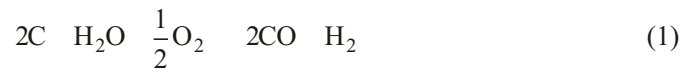
In reality, the synthesis gas contains also CO_2 and CH_4. Since coal gasification is an endothermic reaction, part of the coal is burned to supply the heat needed and to produce CO and CO_2. In the conventional process coal enters the reactor, which operates at a temperature above 1273 K [3]:

In order to produce a carbon-free or a low-carbon fuel gas (mainly H_2), the water gas shift reaction should take place. According to this reaction, CO is converted into CO_2 and H_2. This reaction is exothermic. In the standard process, the syngas enters another reactor with operating temperature below 673 K [3]:

$$CO \quad H_2O \quad CO_2 \quad H_2 \qquad (2)$$

The CO_2 can be captured from the produced fuel gas by using lime sorbent (CaO). This reaction, which is also exothermic, occurs at about 1100 K and produces $CaCO_3$ [3]:

$$CaO \quad CO_2 \quad CaCO_3 \qquad (3)$$

The next step is the regeneration of $CaCO_3$ to release the captured CO_2 and to produce CaO, which will be consequently reused to remove the CO_2 from the fuel gas.

According to the technology described in this paper, the above-mentioned reactions take place in a single reactor. This process is being integrated in a new concept for power plants for the production of H_2 from carbonaceous fuels. The overall reaction is exothermic and, as a result, there is no requirement for direct coal combustion within the gasifier to produce heat for the coal gasification process. The fuel gas mixture that is produced by the integration of the reactions has a high hydrogen concentration. The basic flow sheet of the process is illustrated in fig. 1.
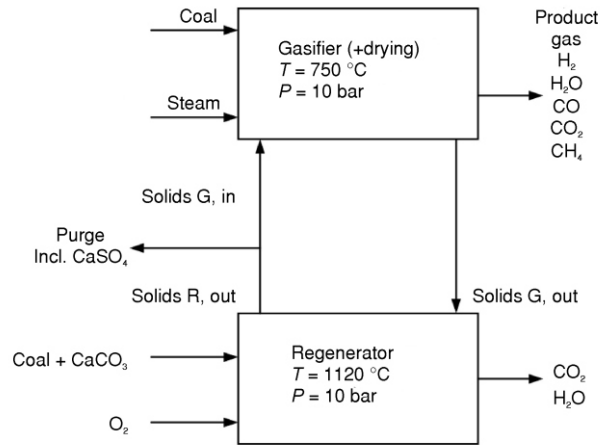
Figure 1. Process flow sheet

Two fluidized beds can be used as the two main process units, namely the gasifier and the regenerator [4, 5, 6]. The operating pressure of both reactors is 10 bar, while the temperature is 750 °C and 1120 °C, respectively. In the calcinator, the reverse calcination occurs at higher temperatures and the sorbent is regenerated. Due to the exothermic CO_2 absorption by CaO, the overall reaction in the gasifier can be adjusted to be slightly exothermic, resulting in minimized energetic losses. The produced fuel gas is hydrogen rich, while the regenerator product gas consists mainly of CO_2 and H_2O. The solids that exit the gasifier are CaO, limestone, lignite ash, CaS, gypsum, and char (C that has not reacted in the gasifier). On the other hand, the solids that exit the regenerator are CaO, ash, and gypsum. The feasibility of the process has been demonstrated by the experimental work conducted for the CO_2 acceptor gasification process [6], the only difference being that they did not produce pure CO_2.

Extra energy is required for the regenerator operation, which is provided by the combustion of the char exiting the gasifier plus additional fuel. This energy includes the sensible heat required from the solids that exit the gasifier at 750 °C and enter the regenerator, which operates at 1120 °C plus the reaction heat of the $CaCO_3$ (make-up $CaCO_3$ plus $CaCO_3$ in the gasifier outlet solids). For the combustion of char and fuel in the regenerator, pure O_2 instead of air is used, thus avoiding air nitrogen. As a result, the regenerator product gas contains mainly CO_2 and H_2O and is ready to be compressed for transportation and storage after being cooled down to near ambient temperature and passed through solids removal filters. Cryogenic air separation is used as the most suitable commercially available technology, a process capable of producing high flows of high-purity oxygen [7].

One significant drawback of the procedure is argued to be the rapid reduction of the CO_2-capture capacity of CaO after a number of cycles, due to difficulties in its continuous reactivation. The calcination process results in the loss of the suitable pore volume in the lime-based sorbent, reducing significantly the carbonation capacity [8, 9]. Due to the absorbent decay, a flow of solids from the outlet of the regenerator should exit the system. The CaO that leaves the system by the purge stream is compensated by the make-up $CaCO_3$ flow in the regenerator.

Simulation of combined cycle fired with the low-C fuel

In order to model the novel concept, a commercial natural-gas fired single-shaft combined cycle (CC) plant of 400 MW net electric power production has been used as the backbone of the simulation. The cycle consists of one gas turbine, one steam turbine and a triple-pressure heat recovery steam generator. The gas turbine exhaust gas enters the Heat recovery steam generator (HRSG), which operates at three pressure water/steam levels (HP, IP and LP) with three drums respectively. Steam from the high-pressure superheaters (HP SH) enters the HP steam turbine (HP ST). From the outlet of the HP ST, it is mixed with steam from the IP SH and enters the boiler for reheating. Consequently, it is admitted to the IP ST. The steam from the outlet of the IP ST is mixed with steam from the LP SH and enters the LP ST. Finally, from the outlet of the LP ST, the steam enters the condenser.

The analysis of the lignite used in the current study is shown in tab. 1. The composition and the LHV of the low-carbon fuel gas produced by the process are shown in tab. 2, as calculated for the fuel presented in tab. 1, assuming equilibrium of the dual fluidised bed process.

Table 1. Lignite analysis

Water [w. %]	50.9
Ash [w. %]	6.77
C [w. %]	29.2
H [w. %]	2.00
N [w. %]	0.47
O [w. %]	9.44
S [w. %]	1.22
LHV [kJ/kg]	9785

Table 2. Fuel gas composition

H_2 [vol. %]	65.81
CH_4 [vol. %]	12.94
CO_2 [vol. %]	0.98
CO [vol. %]	3.21
H_2O [vol. %]	15.73
N_2 [vol. %]	1.33
LHV [kJ/kg]	35419

The equilibrium-based mass balance of the dual fluidized bed process for the production of a low-C fuel is based on the following assumptions:

Gasifier

–   the gasifier operates at 10 bar, 750 °C,
–   char entering the regenerator is 20% of the carbon input to the gasifier,
–   due to the high lignite moisture content, no additional steam is required for gasification, and
–   lignite is not pre-dried before entering the gasifier.

Regenerator

–   the regenerator operates at 10 bar, 1120 °C,
–   char and coal are combusted with pure oxygen produced by a cryogenic ASU,
–   the excess oxygen for char and fuel combustion is assumed to be 2%,
–   fuel and char combustion efficiencies are assumed to be 100%,
–   the fresh limestone and the oxygen are preheated up to 600 °C by the product gas of the regenerator (1120 °C) and the fuel gas (750 °C),
–   the purge rate is 12.7% (Purge flow / Solids R, out flow), and
–   it is assumed for the basic examination of the system described in this work that during gasification, sulphur totally reacts with CaO to produce CaS. In reality, due to the fact that the Ca/S mol ratio is high in both gasifier and regenerator, the above-mentioned reaction is limited only by equilibrium. Additionally, the CaS formed in the gasifier will be oxidised to $CaSO_4$ due to the presence of excess oxygen in the regenerator, whereas the absorption efficiency of SO_2 by the CaO sorbent is enhanced in pressurised conditions. Experimental studies have proven that almost all the sulphur released in the gasifier can be absorbed by CaO to produce CaS. Sulphur rejection of gypsum in the regenerator cannot occur in excess oxygen environments, due to the low dissociation pressure of SO_2 over $CaSO_4$ [6]. As a result, it is assumed in this work that the whole amount of sulphur leaves the system in the form of $CaSO_4$ with the purge flow. Concerning the behaviour of lime at the regenerator conditions with respect to sulphur capture, there are some remaining problems to be solved, which is part of the remaining study to be done.

For the production of 19.4 kg/s of fuel gas, which is the fuel flow required for the simulated CC power plant, 71.1 kg/s of lignite enter the gasifier, corresponding to 20.7 kg/s of carbon. 20% of the carbon content does not react and enters the regenerator (Solids G, out flow in fig. 1). The char flow and an extra lignite flow of 21.9 kg/s are combusted with 31.8 kg/s of pure oxygen, produced by the cryogenic ASU.

The simulation of the CC fired with the low-carbon fuel gas produced from lignite gasification is based on the natural gas fired plant described previously. It is assumed that the water/steam cycle and the fuel heat input remain the same as in the original case. In addition, the flue gas flow entering the turbine is kept the same as in the original cycle. The main results of the simulations are illustrated in tab. 3, where the net efficiency is based on the fuel gas heat input and not on the heat input of the lignite consumed for production of the fuel gas.

Table 3. Simulation results of low-C fuel gas fired CC power plant

Power & Efficiency	Combined cycle fired with low-C fuel gas
GT power output [MW]	252.1
ST power output [MW]	138.2
Block net electric output [MW]	388.5
Block net efficiency [%]	56.5

Results of the process

The block net efficiency of the CC power plant coupled with the system for production of the carbon-free fuel gas is affected by the following power consuming processes:

– lignite gasification and CO-shift reaction. The lignite heat input to the gasifier produces the low-carbon fuel gas, which has a slightly lower energy content. In the case studied, 1 kg of lignite with an LHV of 9785 kJ/kg produces 0.273 kg of fuel gas, with an LHV of 35419 kJ/kg, which equals to 9670 kJ. Due to the high H_2 concentration of the fuel gas, a diluent should be added in order to limit the flame temperature and, consequently, the NO_x production. In the present analysis, however, no NO_x control has been accounted for,
– compression of the low-C fuel gas to 30 bar, in order to be supplied to the gas turbine,
– fuel required for the regeneration process. For 1 kg of lignite entering the gasifier, about 0.31 kg of lignite is consumed in the calcinator to cover the regeneration needs,
– compression of the regenerator product gas up to 110 bar for transportation and storage,
– air separation for the production of oxygen for the nitrogen-free combustion of fuel and char in the calcinator,
– oxygen compression up to 10 bar to enter the regenerator, and
– water pumping for intermediate cooling during the compression of CO_2 and the air that enters the Air separation unit.

Regarding the case studied, the fuel gas heat input of the power plant is 687.1 MW. Consequently, the low-carbon fuel gas consumption is 19.4 kg/s. The lignite input to the gasifier is 71.1 kg/s, which equals to 695.3 MW. In addition, 21.9 kg/s of lignite are consumed by the $CaCO_3$ regeneration process. This equals to a total (gasifier and calcinator) lignite consumption of 909.9 MW.

The compression of the fuel gas from 10 to 30 bar occurs in one step. The fuel gas is assumed to be at 20 ºC and the compressor's isentropic efficiency is 0.85. The power consumed for this process is 1.3 MW.

As far as the CO_2 compression is concerned, three steps with intermediate cooling are required: 10-30 bar, 30-58 bar, and 58-110 bar. The fact that the CO_2 rich gas exits the regenerator at 10 bar pressure decreases considerably the CO_2 compressors power consumption. The CO_2 is cooled before each step to 20 °C and the condensed water is re-

moved. At 58 bar pressure and 20 °C the CO_2 is in the liquid state [9]. The isentropic efficiency ratio of the CO_2 compressors is assumed to be 0.85. The compression power consumed is 19.3 MW.

It is proposed that in order CO_2 of transportation quality, almost all the water content in the gas should be removed due to corrosion issues and the possibility of hydrate precipitation, which can block the transport pipelines. Water condensation with cooling water is not enough, since not all water in the gas is removed. At the pressure level of 30 bar, the gas enters the TEG unit (tri-ethylene-glycol), where the remaining water is almost completely absorbed [10].

Cryogenic air separation can provide high purity oxygen in a large scale. Even oxygen purities of 99.7% are available with this technology. In the case studied, pure O_2 is assumed to enter the regenerator for the combustion of char and lignite. As a result, the ASU power consumption calculations are made for the 99.7% purity case. For 32.5 kg/s of O_2 (81307.4 Nm^3/h) that are required for the combustion of char and lignite with 2% O_2 excess, 142.9 kg/s (402689.7 Nm^3/h) of air enter the Air separation unit, at 5.85 bar, which is the pressure of the distillation column for the 99.7% O_2 purity case [7]. The compression from the ambient pressure up to 5.85 bar occurs in 4 steps with intermediate cooling: 1-1.56 bar, 1.56-2.42 bar, 2.42-3.76 bar, and 3.76-5.85 bar. Air is cooled after each step to 20 °C. The isentropic efficiency ratio of the air compressors is assumed to be 0.83. The compression power that is consumed is about 27.4 MW.

The oxygen exits the ASU at near atmospheric pressure and 18 °C. As a result, extra power is consumed for compression up to 10 bar to enter the regenerator. To compress 32.5 kg of O_2, the power required is 9.9 MW, for a compressors isentropic efficiency of 0.83.

For intermediate cooling during the compression of CO_2 and the compression of air before the ASU, the cooling water is assumed to be at 2.5 bar and the cooling water

Table 4. Novel concept with lime CO_2 capture – Simulation results

Lignite heat input (Gasifier and Regenerator)[MW]	909.9
Compression of the fuel gas in order to enter the GT [MW]	1.3
CO_2 compression [MW]	19.3
ASU [MW]	27.4
Cooling water pumping for CO_2 and ASU air [MW]	0.8
Oxygen compression [MW]	9.9
Net power output [MW]	329.7
Net efficiency [%]	36.2
C entering the gasifier and regenerator (lignite) [mol/s]	2262.8
CO_2 in the flue gas [mol/s]	431.8
C removal efficiency [%]	80.9
CO_2 emissions [kg/MWh_e]	200

pumps power consumption is taken into account for the calculations. The cooling water pumps power consumption for CO_2 intermediate cooling is 0.4 MW, while for the air intermediate cooling it is 0.3 MW.

The simulation results for the CC system coupled with the low-carbon fuel gas production system are illustrated in tab. 4.

Discussion of physical absorption prsocess effect on IGCC efficiency

The coal gasification technology is applied in IGCC systems. Coal gasification processes usually operate with pure oxygen and water vapour and the synthesis gas contains CO and H_2. As mentioned, in order to precipitate the CO_2 before the combustion of the synthesis gas, the CO is converted into CO_2 and H_2 through an exothermic reaction known as "CO shift" or "water shift" reaction. After conversion, the synthesis gas consists mainly of H_2 and CO_2. The advantage of these systems regarding the CO_2 sequestration concept is that the gas volume to be treated is small while the CO_2 partial pressure is high enough, allowing the physical absorption CO_2 capture technique to be an effective and efficient alternative [11].

According to the physical adsorption technology, carbon dioxide is adsorbed physically in a solvent. Henry's law dominates this process, which means that the absorption is pressure and temperature dependent. The CO_2 capture have better results in high partial pressures and low temperatures allowing high levels of separation with high
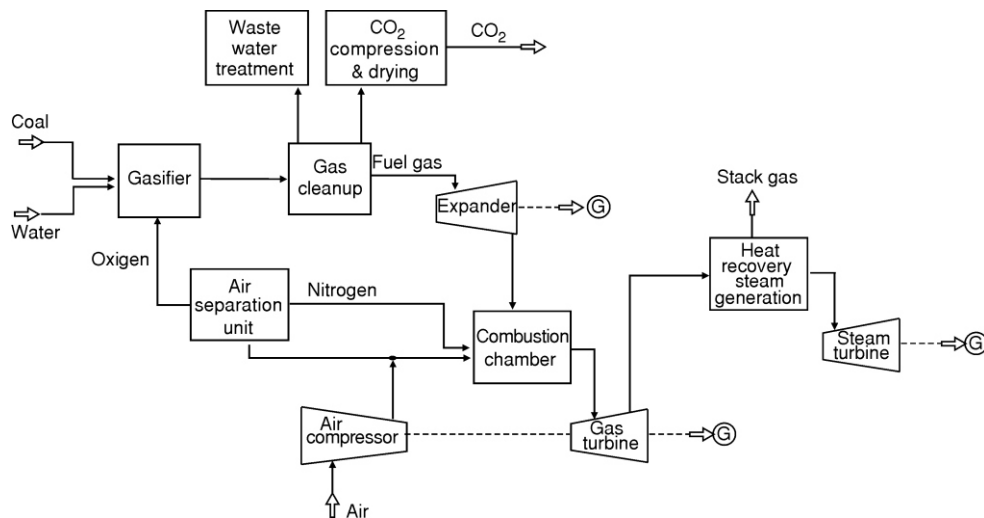

Figure 2. Flow sheet of an IGCC power plant with physical absorption CO_2 capture (source IEA GHG)

purities of CO_2 [11]. The amount of energy required is low. Physical absorbents that can be used for the CO_2 removal from the produced fuel gas are:
– methanol – Rectisol,
– n-methyl-2 pyrrolidon (NMP),
– dimethyl-ether-polyethylene-glycol (DMPEG) – Selexol, and
– propylene carbonate.

Figure 2 illustrates an IGCC plant with CO_2 removal by means of physical absorption.

In tab. 5, the efficiency penalty of a coal fired IGCC power plant with pre-combustion CO_2 capture is illustrated [12]. The CO_2 capture system includes a shift reactor and physical absorption with Selexol. The oxygen required for the process is produced by a cryogenic ASU. The CO_2 is assumed to be compressed up to 110 bar for transportation and storage.

Table 5. Efficiency decrease of IGCC with CO_2 physical absorption

	IGCC original	IGCC with CO_2 capture
Plant size [MW$_e$]	776	676
Net efficiency [%]	43	34
CO_2 emissions [kg/MWh$_e$]	763	142

Preliminary economic considerations

The specific investment cost of an IGCC system without CO_2 capture has been estimated to about 1370 EUR/kW$_e$ [11]. The specific investment cost includes the gasifier, the Air separation unit, the gas cleaning system and the CC unit. The gas cleaning system includes the cyclones and/or filters for separation of the product gas from gasifier solids and the desulphurisation process so that, finally, only clean gas and sulphur leave the gasification plant [14]. On the other hand, the specific investment of an IGCC power plant with physical absorption for carbon dioxide removal is estimated at 1860 EUR/kW$_e$ [12]. This cost includes the following:
– coal gasifier,
– CO shift reactor,
– cryogenic Air separation unit,
– fuel gas cleaning unit, which includes the desulphurisation process,
– CO_2 separation unit with Selexol absorption,
– CO_2 treatment equipment, which includes the CO_2 compressors, the CO_2 cooling systems for water condensation and the TEG dehydration unit, and
– the CC unit.

Regarding the preliminary assessment of the capital requirement of a novel CC integrated with the CaO sorbent CO_2 capture process presented in this paper, the following process units should be considered:

– gasifier (one reactor for the coal gasification, the CO shift reaction and the CO_2 capture from the CaO sorbent),
– regenerator,
– cryogenic Air separation unit,
– fuel gas cleaning system, which would include the removal of the gasifier solids from the fuel gas but not a desulphurisation process,
– CO_2 treatment equipment, which includes the CO_2 compressors, the CO_2 cooling systems for water condensation and the TEG dehydration unit, and
– the CC unit.

Both the novel CC integrating the CaO sorbent CO_2 capture process and the IGCC with CO_2 capture by physical absorption, should include fuel gas treatment systems. According to the above mentioned analysis on sulphur removal, it is expected that no desulphurisation process will be required (however further investigation is required regarding the conditions under which the sulphur removal is efficient) and the investment for the fuel gas cleaning section will be reduced. On the other hand, two reactors are required to perform the gasification, the CO shift reaction, the CO_2 removal and the $CaCO_3$ regeneration. Compared to the IGCC with Selexol CO_2 capture, which needs two reactors (one for the gasification and one for the CO shift) plus the Selexol unit, it is thus expected that the novel concept will not have higher investment costs.

Regarding the variable costs of the novel concept, it should be noted that the make up $CaCO_3$ that should be added in the regenerator due to the decay of the sorbent should also be added to the electricity costs. However, taking into account the significantly higher efficiency of the power plant, the electricity cost of the novel concept with lime CO_2 capture is expected to be lower than that of the IGCC with Selexol CO_2 removal. Finally, as far as the CO_2 avoidance cost is concerned, it is estimated to be about 23.1 EUR/t CO_2 avoided (not including transportation and storage costs) for the Selexol case [12], while for the carbonation/calcination novel concept presented here, there are significant arguments that this cost will be reduced.

Conclusions

According to the preliminary study presented in this paper, the IGCC system with CO_2 capture during coal gasification with CaO is expected to be beneficial in terms of capital investment and electric efficiency compared to a conventional IGCC system with CO_2 physical absorption. The integration of coal gasification, CO shift reaction and CO_2 capture in one single reactor results in an optimized low-carbon fuel gas production with decreased energy and efficiency penalties. However, due to the fact that this pre-combustion concept is new, further investigation should be conducted regarding the process flow and process units of such a system. Further considerations of the IGCC with

integrated gasification/CO_2 removal process include the thermodynamic optimization as, for example, the recovery of the high energy content of the fuel gas that exits the gasifier and the waste gas that exits the regenerator. Taking into account that both streams should be cooled down to near ambient temperature, the sensible heat could be integrated in the water/steam cycle of the CC power plant.

Acknowledgments

Abbreviations

ASU   – air separation unit
CC   – combined cycle
G   – gasifier
GT   – gas turbine
HP   – high pressure
HRSG – heat recovery steam generator
IGCC – integrated gasification combined cycle
IP   – intermediate pressure
LHV – lower heating value
LP   – low pressure
NG   – natural gas
R   – regenerator
RH   – reheater
SH   – superheater
ST   – steam turbine
TEG – tri- ethylene- glycol

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