

Carbon Dioxide Capture for Storage in Deep Geologic Formations – Results from the CO₂ Capture Project

**Capture and Separation of Carbon Dioxide
from Combustion Sources**

Edited by

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Chapter 12

GENERATION OF HYDROGEN FUELS FOR A THERMAL POWER PLANT WITH INTEGRATED CO₂-CAPTURE USING A CaO–CaCO₃ CYCLE

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ABSTRACT

A new integrated reforming reaction for hydrogen production is simulated. Hydrogen gas is produced from natural gas and water in a modified reforming reaction where CO₂ reacts with a metal oxide (MetO, e.g. CaO) to form a metal carbonate (MetCO₃, e.g. CaCO₃). The carbonate is decomposed thermally in a separate reaction and the metal oxide is recycled back to the reformer. This provides an efficient means of separation of the carbon dioxide from the reformer. The exothermic carbonation reaction provides most of the energy necessary to drive the hydrogen-producing reaction to completion. The CO₂ removal process has been designed and simulated to test the generation of hydrogen fuels for a thermal power plant. Although, the concept originally was intended for integration with processes with high-temperature waste heat, the thermodynamic analysis shows that the process can be used for hydrogen production for a combined cycle power plant and steam boilers as well.

INTRODUCTION

Conversion of natural gas and other light hydrocarbons via steam reforming is currently the major process for hydrogen production and will probably remain the process of choice for the next few decades. However, this process involves multiple steps and severe operating conditions. The primary reformer operates at approximately 800–850 °C and 20 bar and large quantities of fuel must be burned to supply the energy necessary to maintain the reformer temperature. Moreover, the process requires three other steps: two shift reactions (high- and low-temperature) and a CO₂-separation step, often an amine scrubbing process [1].

Within the last few years, the concept of combining reaction and separation to simplify chemical processes, conserve energy, and/or to improve product quality and yield has received increased attention. The addition of a CaO-based sorbent to selectively remove CO₂ in synthesis gas applications is one example. Hydrogen gas can be produced by passing a steam–methane feed over a mixture of reforming catalyst and CO₂-sorbent, and removal of CO₂ as it is formed, allows the reforming and shift reactions to proceed almost to completion in one single step [2].

In a research project run by Institute for Energy Technology (IFE) and the Christian Michelsen group (CMR/Prototech AS), a new integrated reforming reaction for hydrogen production is being developed. Hydrogen gas is produced from natural gas and water in a modified reforming reaction where CO₂ reacts with a metal oxide (MetO, e.g. CaO) to form a metal carbonate (MetCO₃, e.g. CaCO₃). The carbonate is decomposed thermally in a separate reaction and the metal oxide is recycled back to the reformer. This provides an efficient means of separation of the carbon dioxide from the reformer. The exothermic carbonation reaction provides most of the energy necessary to drive the hydrogen producing reaction to completion. Figure 1 shows the concept schematically and illustrates how high-temperature waste heat from a solid oxide fuel cell for electricity production based on natural gas can be utilised.

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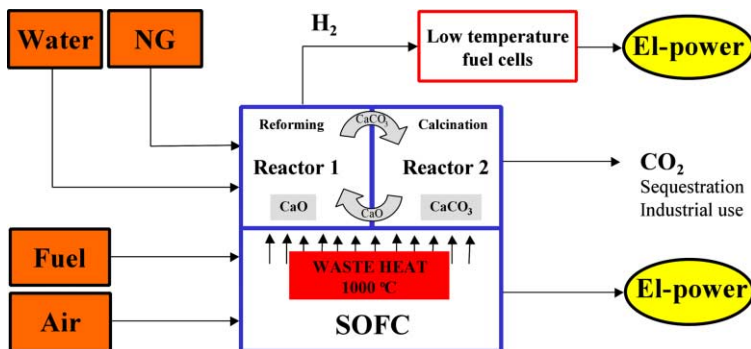


Figure 1: A schematic drawing for a possible use of waste heat from an SOFC for the production of hydrogen and electrical power. Reactor 1 is for the hydrogen production and CO₂ capture as, e.g. CaCO₃ ($\text{CH}_4 + 2\text{H}_2\text{O} + \text{CaO} \leftrightarrow 4\text{H}_2 + \text{CaCO}_3$). Reactor 2 is used for the calcination reaction ($\text{CaCO}_3 \leftrightarrow \text{CaO} + \text{CO}_2$), where the CO₂-sorbent is regenerated.

The SOFC can be fed with pre-reformed natural gas (syngas) or, in a more integrated system, with a fraction of the hydrogen stream produced in the reforming reaction.

The main advantages of such a hydrogen-production process are:

1. Process simplification: reforming, water gas shift and CO₂ separation occur simultaneously in the same reactor.
2. Increased hydrogen yield: high H₂ yield at lower temperatures than in the conventional reforming process.
3. Separation of CO₂ as a solid in the process: no additional costly step for CO₂ separation. The CO₂ is delivered as a pressurised, concentrated CO₂ stream ready for sequestration or utilisation.
4. Recycled CaO represents an important carrier of heat into the reforming stage.

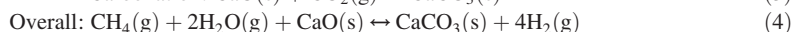
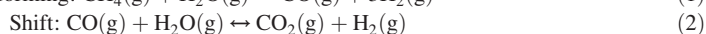
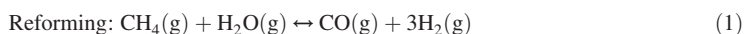
In the present work, it is suggested and described how such a CO₂ removal process can be integrated in a hydrogen combined cycle power plant (CCPP) and a hydrogen steam boiler, that is, generation of a N₂-diluted H₂ fuel gas stream, (H₂/N₂ ratio: 50/50) and generation of a hydrogen-rich fuel (+95%).

High-temperature waste heat is not available in a CCPP and a hydrogen steam boiler and it is therefore necessary to establish a suitable technological interface between the CO₂-capture cycle and the hydrogen CCPP and steam boiler application technologies.

STUDY METHODOLOGY

Thermodynamic and Process Analysis

Conventional steam reforming is a multiple step process with steam reforming (1) and water gas shift (2) reactions. When utilising the carbonation reaction by adding an absorbent such as CaO (3), all three reactions occur simultaneously and allow for hydrogen production in one single step at the same time as CO₂ is separated as a solid in the process (4). In addition, when the CO₂ gas is removed in the process, the equilibrium in the reaction is shifted towards higher hydrogen yield.



Both in the present concept for hydrogen generation (4) and in the conventional steam reforming reactions (1) and (2), maximum hydrogen yields can be calculated assuming thermodynamic equilibrium. Thermodynamic calculations are shown in Figure 2 which compares the H_2 yield in the product gas as a function of temperature at 5 bar and a steam to methane ratio (S/C) of 2 and 4, respectively.

In the conventional reforming process (without CaO), the hydrogen content increases with increasing temperature and reaches a maximum of about 76% at 950 °C for a steam to methane ratio of 2 and about 77% at 850 °C for steam to methane ratio of 4. This increase is governed by the endothermic reforming reaction. On the other hand, with CaO, the hydrogen content reaches a maximum of about 90% at 685 °C for a steam to methane ratio of 2, and about 97% at 650 °C for a steam to methane ratio of 4 (in both cases the formation of $Ca(OH)_2$ has been taken into account). At lower temperatures, essentially all of the carbon oxides are removed by the sorbent and the major impurity in the hydrogen gas is CH_4 . At higher temperatures, more CH_4 is converted and the main impurities are CO and CO_2 . Without the CO_2 -sorbent, a reformer temperature of about 825 °C would be required to achieve the maximum H_2 content (followed by shift reaction and CO_2 separation) comparable to what can be achieved with the CO_2 sorbent system at about 650 °C.

A simple energy balance calculation based on the above assumptions favours slightly the present concept compared to a conventional steam reforming process, 240 kJ/mol CH_4 and 250 kJ/mol CH_4 , respectively.

Reactor Technology

In order to separate the CO_2 produced in the reforming reaction in a continuous way, the CO_2 sorbent has to be regenerated from the carbonate to the oxide. Two regeneration concepts have been considered:

1. a batch process where two reactors change operating mode, that is each vessel is sequentially operated as a reformer and a regenerator without transferring catalyst and sorbent particles between the vessels, or
2. a continuous process where both vessels are dedicated reactors, which do not change operating mode. Sorbent (and catalyst) particles circulate between the two reactors for regeneration and CO_2 absorption, respectively.

A batch process will require a huge reactor volume to produce an acceptable quantity of hydrogen in each batch, and consequently large amounts of sorbent and catalyst will be necessary. Moreover, reforming and calcination will be difficult to optimise in the same vessel and the two reactions will also be difficult to synchronise. The reactor's atmosphere must be changed between each batch and this implies more steps in the process. Finally, due to the different operating conditions, heating and cooling of the reactors when changing from reformer to regenerator mode will be slow due to the large thermal capacity of the acceptor material.

A continuous process will allow for a more compact reactor design and it will be easier to optimise both reactions (optimal conditions for both reactors at any time and no atmosphere shifting of the reactors). Smaller reactor volumes can be designed and the amount of sorbent can be minimised. The system will also require less auxiliary equipment than the batch system because each reactor is dedicated for its purpose. Finally, as the operating mode does not change, no idling is necessary for heating and cooling of the reactors and the plant regulation can respond faster to load changes. Consequently, a continuous flow process with a circulation of solids between the reactors is chosen for the present case studies.

Possible reactor system

A system very similar to one of the fluid catalytic cracking (FCC)-systems available for industrial production developed by Standard Oil Development Company Inc. (SOD Model IV) could be suggested for the present hydrogen production reaction. It is a two-dense-region circuit composed of two fluidised bed reactors with a pair of U-tubes for circulating the mixture of reforming catalyst and calcium oxide. The gas injection rate into the transfer line controls the rate of solid circulation, and for stable operation the pressure in the two units is kept close to the same value [3]. The reactor system is shown in Figure 3.

This solids circulation system is based on the liquid-like behaviour of fluidised beds. The operating principle for a stable circulation system for solids is shown in Figure 4.

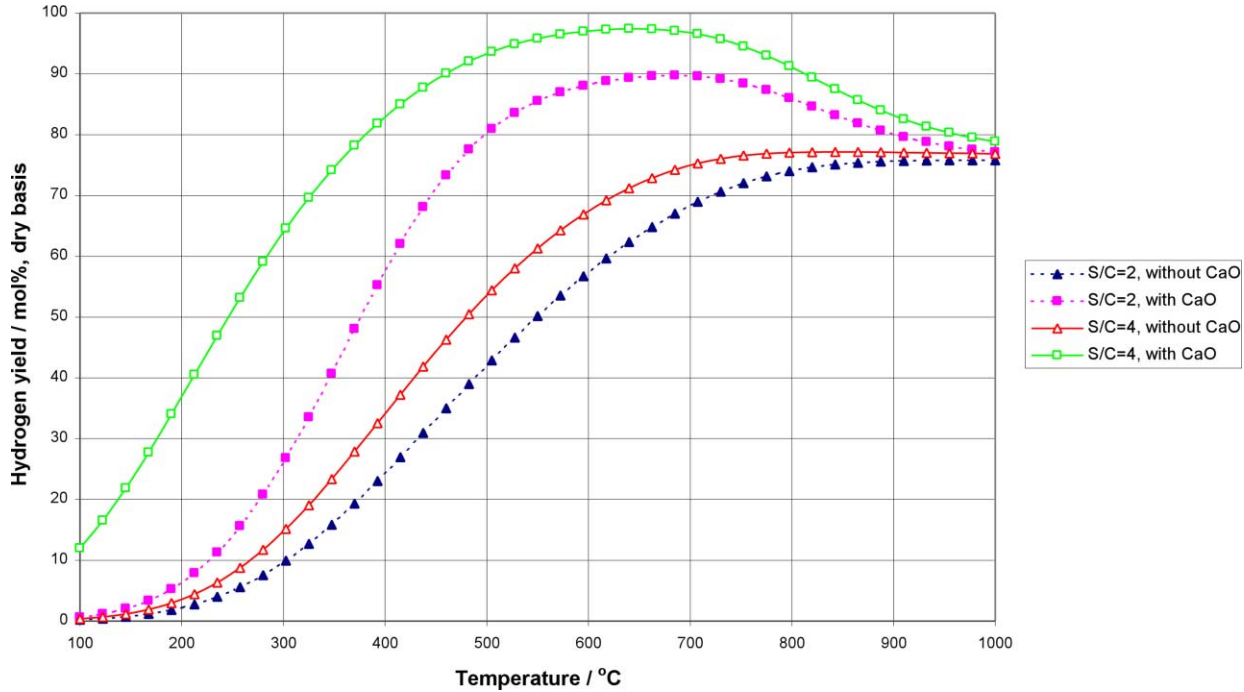


Figure 2: Equilibrium hydrogen content as a function of temperature with and without the CO_2 sorbent. $P = 5$ bar and two different steam to methane ratio ($S/C = 2$ and 4).

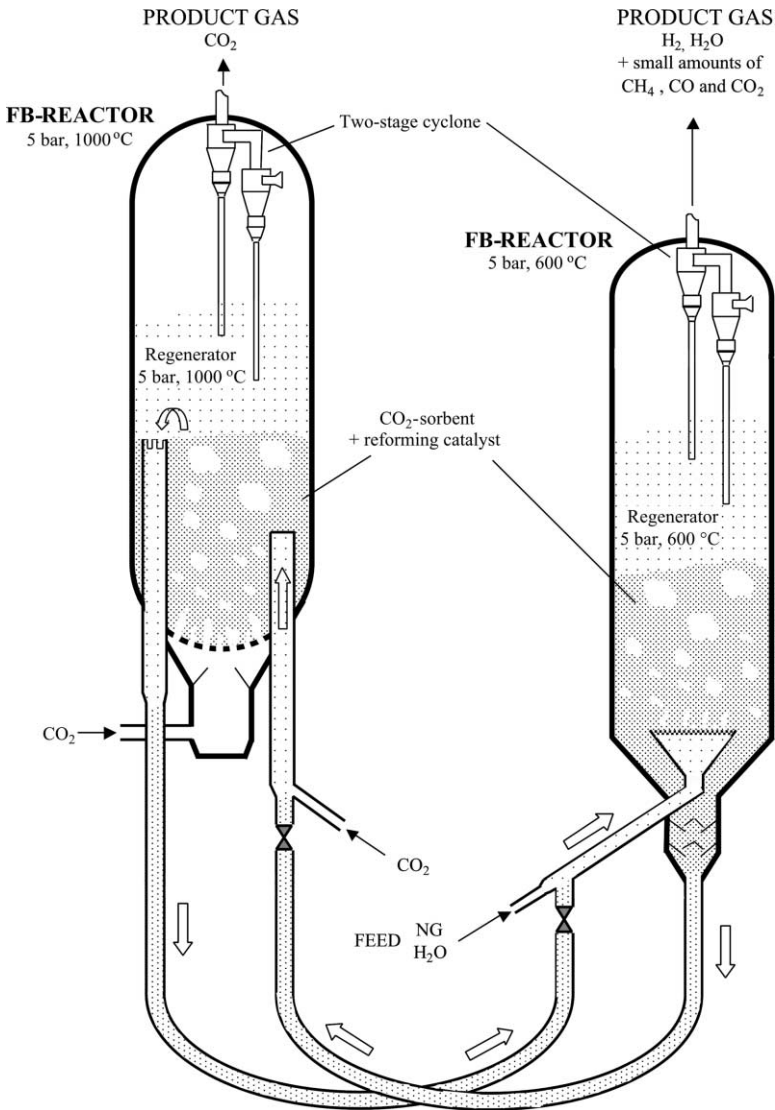


Figure 3: Suggested reactor system for hydrogen production with integrated CO₂ capture. The proposed reactor system is composed of two fluidised bed reactors with a pair of U-tubes for circulating the mixture of reforming catalyst and CO₂ sorbent.

If a gas is injected in pipe C connecting two fluidised beds A and B and if all the contents of beds and pipe are fluidised, then it can easily be shown that the difference in static pressure in the two sides of the pipe will be the driving force causing the particles to flow from A to B. A combination of two such piping arrangements will then give a complete circulation system for the solids. A balance between the frictional

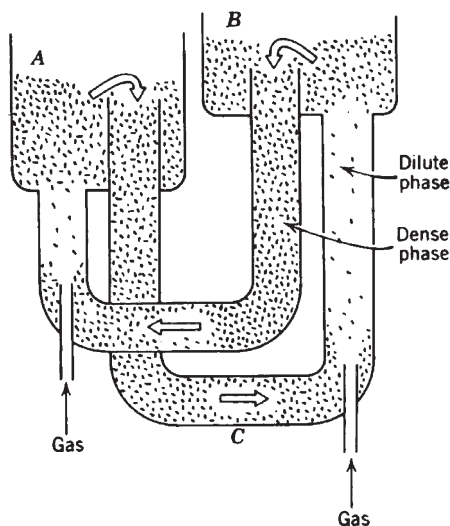


Figure 4: Operating principle for a stable circulation system for solids.

resistance and the previously mentioned pressure differences gives the rate of circulation. The circulation is controlled either by changing the frictional resistance of the system to flow, say, by slide valves or by varying the average densities of the flowing mixture in the various portions of the connecting circuit, a procedure which modifies the pressure differences.

Because of the large specific heat of the solids, their rapid movement between reactor and regenerator can transport large quantities of heat from one to the other, and as a result these circulating solids can be used most effectively to control the temperature of the system [3].

In the present case, the conversion of reactants controls the system. For this reason, both reactors should include internals like baffle plates or horizontal perforated plates for example. In a bed with internals, the bubble size is close to constant, so for a given weight of solids and volumetric gas flow rate the aspect ratio (height-to-diameter ratio) has only a small effect on conversion. As a safe value the minimum aspect ratio should be about unity [3].

Design Basis and Operating Conditions

The different data and parameters used in this study are shown in Table 1, and the operating conditions chosen for the two reactors are listed in Table 2.

RESULTS AND DISCUSSION

Combined Cycle Power Plant System

A description of a possible process design is shown in the process flow diagrams (PFD) in Figures 5 and 6. Desulfurised natural gas at 400 °C and 5.4 bar is fed into the plant. Water at 10 °C and atmospheric pressure, is pressurised and exchanges heat with the compressed CO₂ in order to liquefy the CO₂. The pure CO₂ stream is now ready for high-pressure sequestration. The water is mixed into the natural gas stream before the mixture is heated in three heat exchangers and fed into the reformer. The produced H₂-stream from the reformer is split. About 41% of the gaseous reformer products are fed into the combustion chamber of the regenerator to produce the heat required to regenerate the CO₂ sorbent. The CO₂ sorbent enters the reformer at high temperature (1000 °C) and reacts with the CO₂ produced in the reforming process to yield CaCO₃. The reformer is assumed to have a pre-reforming stage converting all hydrocarbons >C1 to methane,

TABLE 1
DATA AND PARAMETERS USED IN THE STUDY

<i>Desulfurised natural gas feed</i>	
Composition	Mol%
N ₂	0.61
CO ₂	2.92
CH ₄	79.78
C ₂ H ₆	9.68
C ₃ H ₈	4.45
<i>n</i> -C ₄ H ₁₀	1.23
<i>i</i> -C ₄ H ₁₀	0.73
<i>n</i> -C ₅ H ₁₂	0.20
<i>I</i> -C ₅ H ₁₂	0.21
C ₆ H ₁₄	0.21
<i>Ambient conditions</i>	
Average ambient temperature	10 °C
Average sea water temperature	10 °C
<i>Other data</i>	
Average load power station	100%
Power output (net)	330 MW
CCPP efficiency	58%
CO ₂ removal (total)	90%
CO ₂ exit pressure	80 bar
CO ₂ production	1 Mt/year
NO _x emissions	Not investigated

TABLE 2
OPERATING CONDITIONS FOR THE TWO REACTORS

<i>Reformer: hydrogen production/integrated CO₂ capture</i>	
Steam to natural gas ratio	3.5
Calcium oxide to natural gas ratio	1.5
Temperature	600 °C
Pressure	5 bar
<i>Regenerator: calcination reaction</i>	
Temperature	1000 °C
Pressure	5 bar
Atmosphere	CO ₂

hydrogen and carbon dioxide. The exhaust from the regenerator combustion chamber is split in order to dilute the H₂ stream from the reformer to 50% H₂. This mixture is fired into the gas turbine combustion chamber. The exhaust gases from the gas turbine and from the regenerator combustion chamber are mixed and then sent to a high-recovery steam generator (HRSG) coupled to a three stage double reheat steam turbine bottom cycle.

The CCPP process has been simulated on the HYSYS process version 2.4 steady-state process simulator, a software product from Hyprotech AEA Technologies.

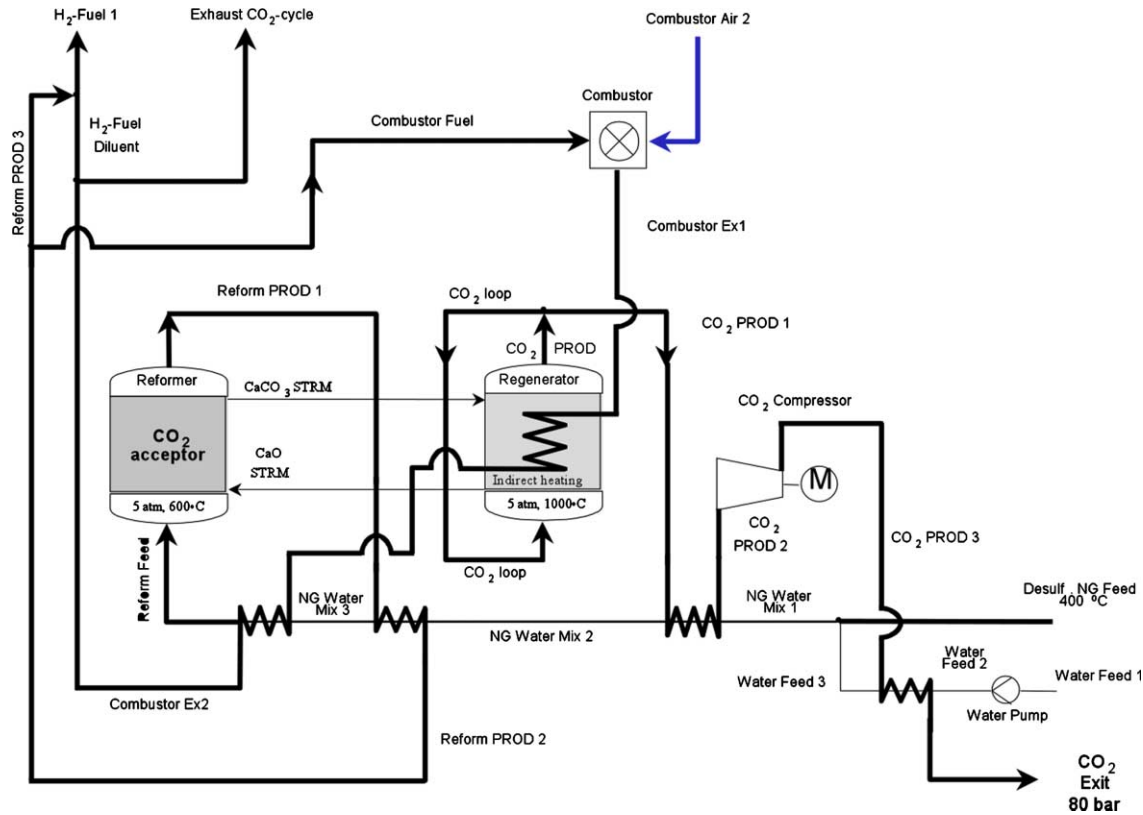


Figure 5: Process flow diagram, IFE CO₂-capture concept, CCPP system.

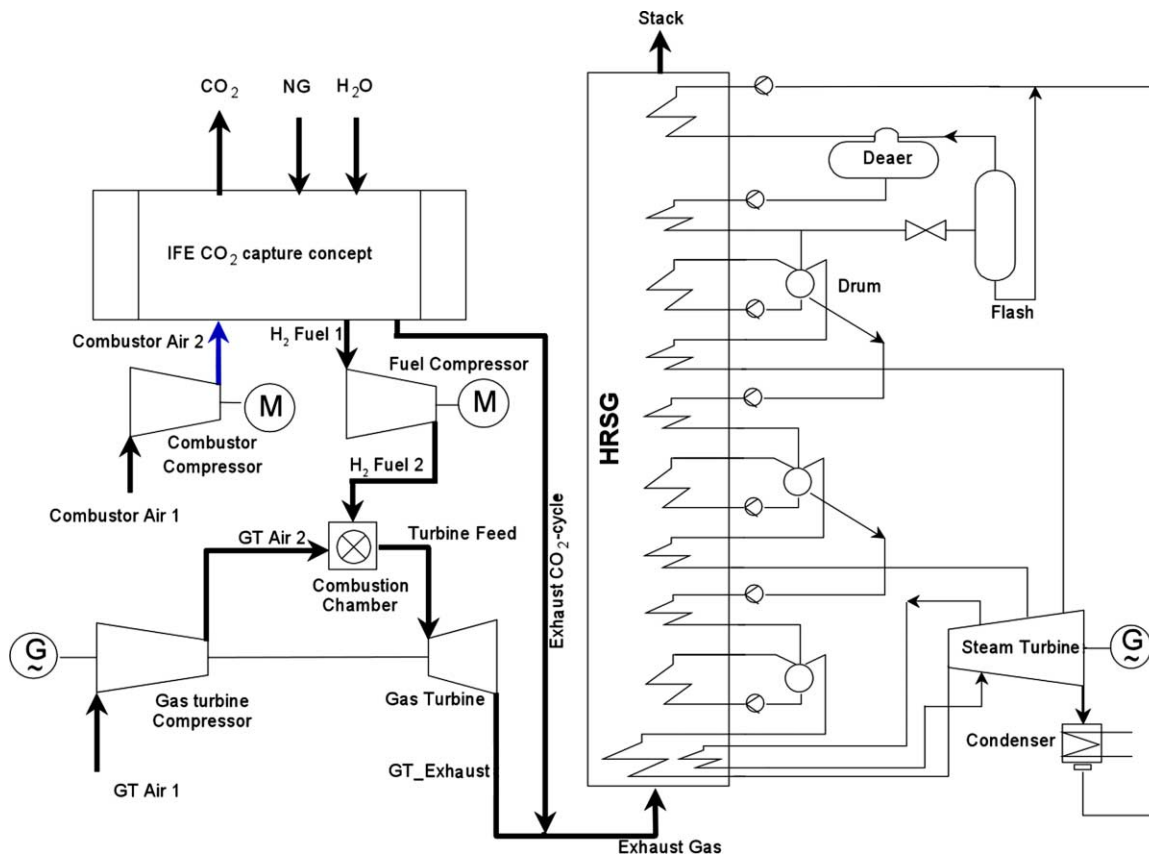


Figure 6: Process flow diagram, overall process, CCPP system.

Production and consumption figures

A summary of production and consumption figures is shown in Figure 7. In general, it is found that the H₂ stream from the reformer contains 0.9% CH₄ and 1.0% CO₂, which goes into the gas turbine combustion chamber and produces CO₂ which again is released to the atmosphere. Given the assumptions described below, the CO₂ capture is 89.9%, thus practically meeting the CO₂ capture requirement of 90%.

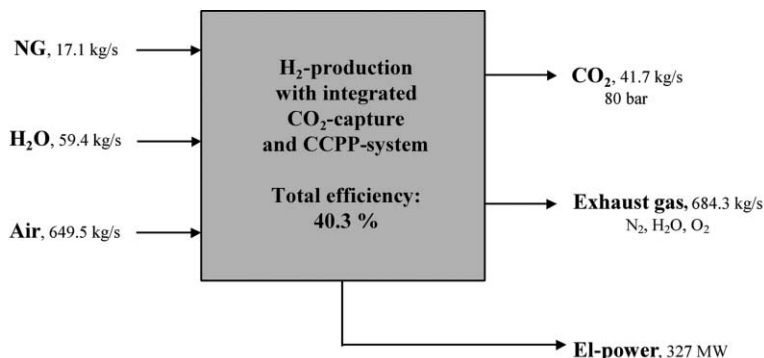


Figure 7: Production and consumption figures, CCPP system.

The natural gas flow is 17.09 kg/s at an LHV of 45.536 MJ/kg. The primary energy going into the process is thus 778.1 MW. The CCPP generates 403.6 MW gross. However, the air compressor and the fuel compressor in the CO₂ removal process consume 23.7 and 32.8 MW, respectively. CO₂ compression to 80 bar also consumes 14.2 MW. Other auxiliary power requirements are assumed to be 3 MW. Net power output is thus 326.8 MW. Taking into account that the desulfurisation and heating of the natural gas to 400 °C, one arrives at an overall LHV electrical efficiency of 40.3% for the CCPP.

Steam Boiler System

A description of the process design is shown in the PFD diagrams in Figures 8 and 9. The CO₂ removal process is basically identical to the CCPP case except that the H₂ product is not diluted as described previously. Instead the H₂ product is cooled to remove the water vapour and meet the purity requirements (>95%). A large amount of excess heat is utilised for steam production as a secondary product.

The same program (HYSYS process) has been used to simulate the process.

Production and consumption figures

A summary of production and consumption figures is shown in the diagram in Figure 10. In general, it is found that the H₂ stream from the reformer contains 0.9% CH₄ and 1.0% CO₂. Given the assumptions described below the CO₂ capture is 89.9%, thus practically meeting the CO₂ capture requirement of 90%.

The natural gas flow is 17.09 kg/s at an LHV of 45.536 MJ/kg. The primary energy going into the process is thus 778.1 MW. However, the air compressor in the CO₂ removal process consumes 8.2 MW and CO₂ compression to 80 bar also consumes 14.2 MW. Other auxiliary power requirements are assumed to be 3 MW. The products are a hydrogen stream with an energy content of 552.3 MW and 97.2 kg/s steam at 106 bar and 289 °C.

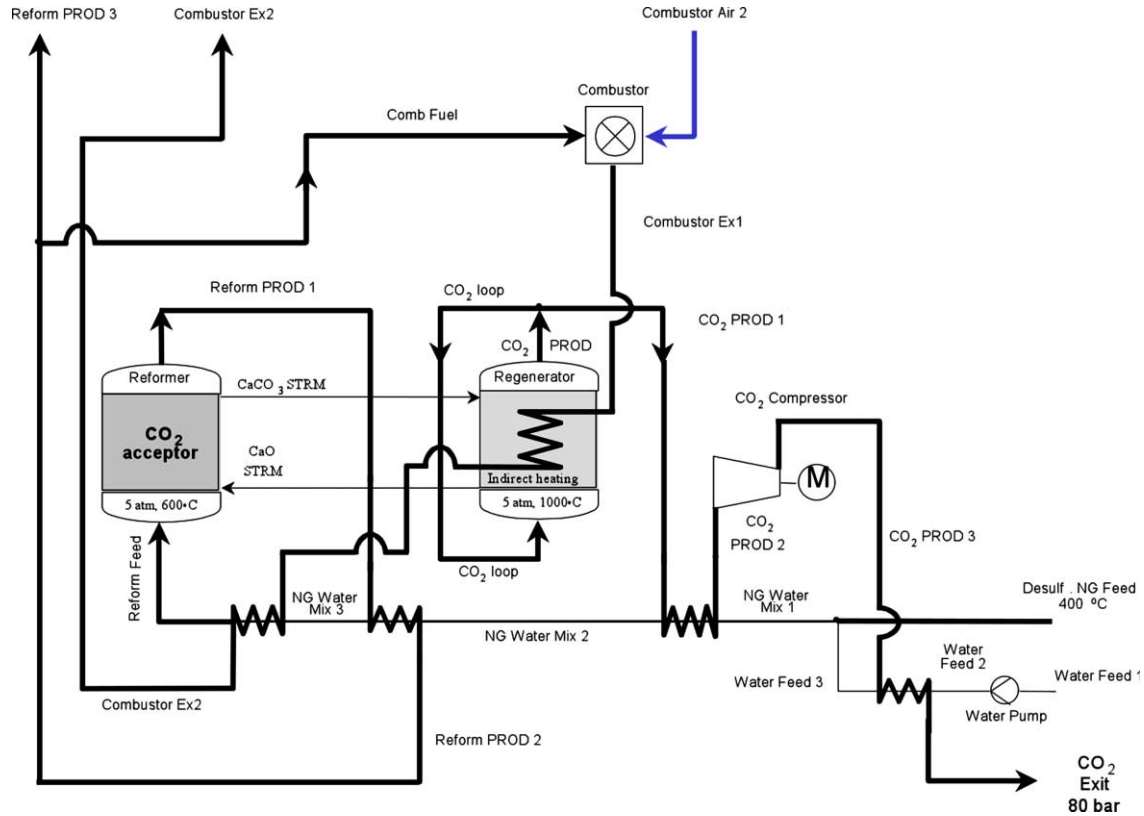


Figure 8: Process flow diagram, IFE CO₂-capture concept, steam boiler system.

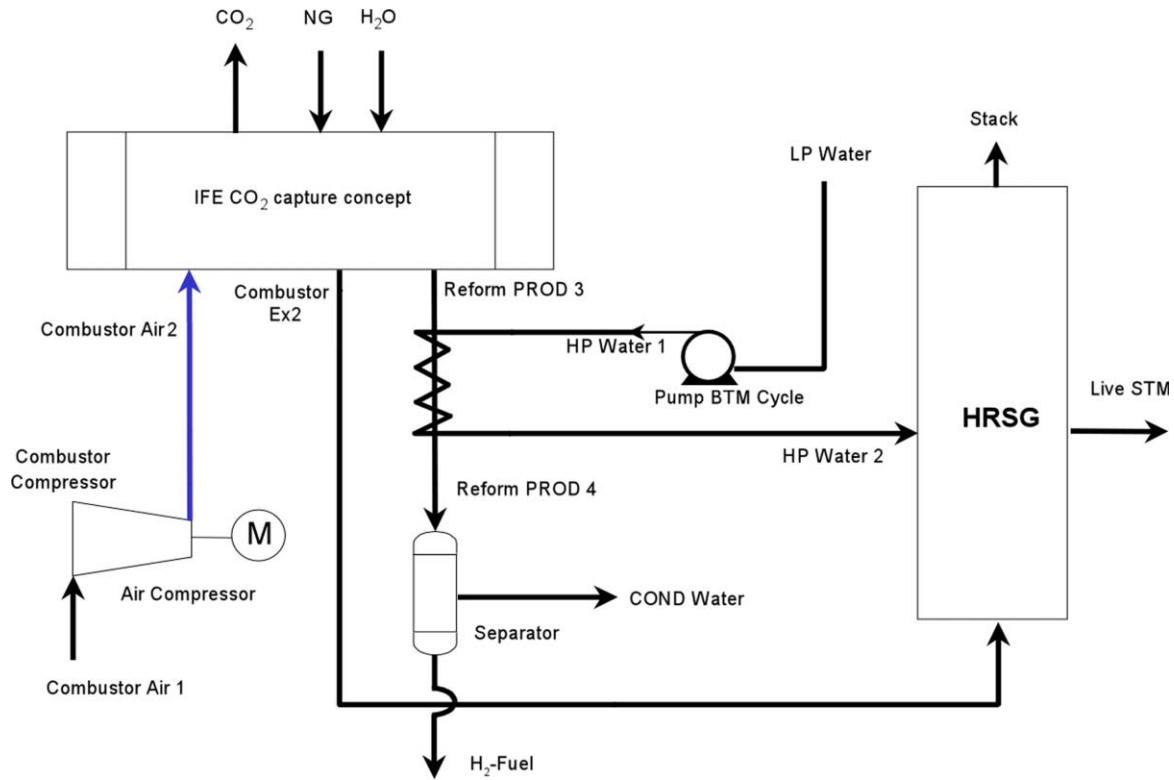


Figure 9: Process flow diagram, overall process, steam boiler system.

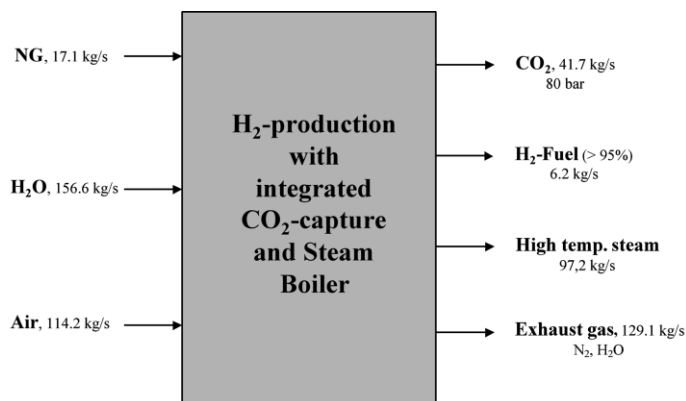


Figure 10: Production and consumption figures, steam boiler system.

Assumptions and Simplifications

The following assumptions and simplifications have been made both for the CCPP and the steam boiler:

1. Desulfurised natural gas is fed into the process. However, the heat from the desulfuriser is brought along producing a natural gas feed at 400 °C and 5.4 bar.
2. The conversion rate of hydrocarbons heavier than methane is 100%.
3. The reformer conversion rate for methane is 93%.
4. The CO₂ absorption rate is 95%.
5. The excess CaO is approximately 50% compared to hydrocarbon C.
6. The CaCO₃ conversion in the calcination reaction is 99%.
7. A catalyst has not been defined in the simulation. Consequently, one assumes that the catalyst is not circulating between the reactors and not affecting the heat balance of the system.
8. The combustion chamber of the regenerator is inside the reaction vessel for maximum heat transfer.
9. The CO₂ loop shown in Figures 5 and 8 is regarded as a reactor-specific process and is not considered in the simulation.
10. The adiabatic compressor efficiencies are assumed to be 85%.
11. Heat loss to the environment from the CO₂ removal process reactors and pipes, etc. is assumed to be 9.3% of the heat transferred in the heat exchangers.
12. The LHV efficiency of the CCPP is 58% equivalent to the value assumed in similar calculations [4].

It is assumed that water vapour in the turbine fuel does not affect the firing properties of the combustion chamber. Instead of 50% dry N₂, the simulation allows for steam in the inert fraction as long as 50% H₂ is maintained.

In the simulations, it is assumed that pure CO₂ exits the regenerator. However, in the proposed circulating fluidised bed system, some minor gas leakages from one reactor to the other have to be considered and will affect slightly the purity of the CO₂ stream. The main pollutants will be small amounts of hydrogen, steam, methane and carbon monoxide. A proper design of the U-pipes and a good control of the circulation flow will minimise these gas leakages.

CONCLUSIONS

Given the above assumptions and additionally assuming that the fluidised bed reactors can be designed to meet the assumptions in real life, the HYSYS simulations show that 90% CO₂ removal is possible using hydrogen produced in a reforming reaction with integrated CO₂ capture based on a CaO–CaCO₃

cycle. Although reactor design is not a part of this pre-study, it should be pointed out that the assumptions are quite demanding on the reactor efficiency, especially because the fluidised beds are one stage reactors.

The HYSYS model simulation showed that a CCPP with an electrical efficiency of 58% (LHV), which was fed with hydrogen from the present integrated reforming reaction can achieve an overall plant efficiency of about 40%. This is somewhat less than comparable CO₂ removal processes. According to Bolland et al. [4], the best-known concepts yield LHV efficiencies in the range 49–51%. The main reason for the lower efficiency in the present concept is that the CO₂ removal process is intended to operate at lower pressures and consequently, the hydrogen fuel has to be compressed before entering the turbine combustion chamber. Another loss factor is the need for sulfur removal. If one can design a continuous process where the reforming reaction is kept at a high pressure (15 bar, for example) while the regeneration process is still at low-pressure (1–5 bar), one can avoid the use of a power consuming fuel compressor. This design will increase the efficiency to 44.3%. Similar calculations with a reduced steam to carbon ratio (2.0 instead of 3.5) and keeping the natural gas feed constant and all reactor pressures and temperatures at the same level, showed that the CO₂ removal drops to about 86% due to a reduced methane conversion in the reformer. Less heat is then needed for CO₂ recovery and steam generation and a larger fraction of the hydrogen produced in the reformer is thus available as CCPP fuel. The net electrical efficiency increases to about 43–44%.

In the present process with the high temperatures and circulation of large amounts of solid material, it seems difficult to reach the same efficiency figures as comparable concepts. Heat losses and auxiliary power demand will inherently be higher in the present concept.

RECOMMENDATIONS

Mitigation of CO₂ emissions from energy production will require new technical solutions. Future energy technology must comply with both the requirement for increased energy and cost efficiency as well as the need for significant reductions in the CO₂ emissions. There is, therefore, a strong need for new technologies with potential possibilities of electricity production at the same costs as conventional power plants, with emphasis on strongly reduced CO₂ emissions. The “hydrogen society” is believed to be one answer to these challenges and in this society fuel cells and gas reforming are expected to play a major role. For CO₂ capture, the work should focus on pre-combustion technologies, as a precursor to the coming hydrogen society. Integration of different novel and radical technologies will be important in order to obtain high total efficiencies, flexible solutions with respect to fuel and market demands as well as competitive costs.

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