Carbon Dioxide Capture for Storage in Deep Geologic Formations – Results from the CO_2 Capture Project

Capture and Separation of Carbon Dioxide from Combustion Sources

Edited by

David C. Thomas
Senior Technical Advisor
Advanced Resources International, Inc.
4603 Clearwater Lane
Naperville, IL, USA

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Chapter 11

PRE-COMBUSTION DECARBONISATION TECHNOLOGY SUMMARY

Henrik Andersen

Norsk Hydro, ASA, Oslo, Norway

ABSTRACT

The CO_2 Capture Project (CCP) was formed in late 2000 and after a review and evaluation phase began actual technical development work near the end of 2001. Most of the technology providers had only 2 years to complete their work. Even then, significant progress and advances in several key areas were made. New insights on adoption of existing technology in the CCP industrial scenarios were achieved. The key results from the pre-combustion technology development projects are:

- Four new advanced technologies were developed to "proof-of-concept" with significant advancement in efficiency, cost and CO_2 capture compared to the best available capture technology.
- The four technologies showed cost reduction potential in the range from 30 to 60%, with the Hydrogen Membrane Reformer demonstrating the highest potential.
- Three of the new advanced technologies were developed for different CCP scenarios. The designs were checked, integrated, and cost estimated by an independent contractor (Fluor) in order to assure design quality and consistency when comparing with the baseline technology, thus enhancing credibility of the conclusions.
- Significant advancements were made in hydrogen membrane materials covering a wide temperature range.
- Further development is needed to advance the most promising technologies, however, it is expected that new technologies can be developed and demonstrated in 2010–2015 with costs in the range of $15–40 MM.
- Pre-combustion technology can be developed to meet stringent requirements on NO_x, CO, and SO_x formation. The lowest NO_x formation was predicted to be 5 ppm vol. from a combined cycle gas turbine. For open-cycle gas turbines, the NO_x formation was reduced by 50%. CO and SO_x formation were virtually zero.
- Pre-combustion technology can be designed as stand-alone facilities for both retrofit and new build applications giving a wide application range and benefits with respect to integration in existing complex facilities, e.g. refineries.
- Pre-combustion technology can be used for other applications, e.g. gas-to-liquids (GTL), ammonia, hydrogen and syngas production, thus increasing the economic potential of the technology and return of investment.
- Significant improvement in energy and CO_2 capture efficiency was obtained for several technologies, resulting in an efficiency penalty for combined cycle gas turbines of less than 5% with nearly 100% CO_2 capture.
- A 15% improvement of gas turbine heat rate can be obtained when switching from natural gas to hydrogen-rich fuel, making the pre-combustion technology a strong candidate for the large numbers of open-cycle gas turbines in operation in the US.
- Demonstrated very low CO_2 avoided costs for the Canadian scenario—CO_2 capture from petroleum coke fired IGCC—approximately $10–15 per ton.
- Existing pre-combustion technology can be considered proven for a wide range of CO_2 capture applications including the CCP scenarios.

INTRODUCTION

The CO_2 Capture Project (CCP) pre-combustion technology development was the largest CO_2 capture program in the CCP. It involved 13 individual projects completed by 20 different technology suppliers.

The studies are divided into three key areas:

- *Membrane studies*: development of hydrogen membranes and reactors for steam reforming and water gas shift application.
- *Enhanced hydrogen production*: novel non-membrane technology for syngas and CO_2 capture application.
- *Integration and scale-up studies*: existing technology integrated and optimised for the CCP scenarios.

All the technologies have been developed to be used in the real-life industrial scenarios defined by the CCP. This approach gave the most insight into the economic potential and technical performance of the technologies.

History of Pre-combustion Technology

Pre-combustion technology is based on well-known technologies that are currently used in commercial operations such as: hydrogen, ammonia and syngas production. The technology comprises two main steps:

reforming/conversion of fossil fuel to syngas (a mixture containing hydrogen, CO, and CO_2), and
separation of CO_2 and hydrogen to produce a hydrogen-rich stream.

Conversion of fossil fuel to syngas dates back to several centuries when coal was the primary energy source. The Scottish engineer William Murdoch was the first to convert fossil fuel to syngas who in 1792 used the syngas to light his house. Syngas was later called “town gas” or “city gas”. “Gas” lighting was widespread between 1800 and 1920 for lighting homes and businesses. In the United States, more than 1000 town gas plants (Figure 1) were in operation in 1905. The technology developed from gasification of coal to reforming of natural gas through use of catalysts. Steam reforming technology, introduced in the 1930s, remains the primary method to convert natural gas to syngas. More than 90% of the present hydrogen production—500 billion m^3 per year according to IEA—is based on reforming of fossil fuel. This volume suggests that about 500 reformers with an average capacity of 100,000 Nm^3/h hydrogen are in operation worldwide.

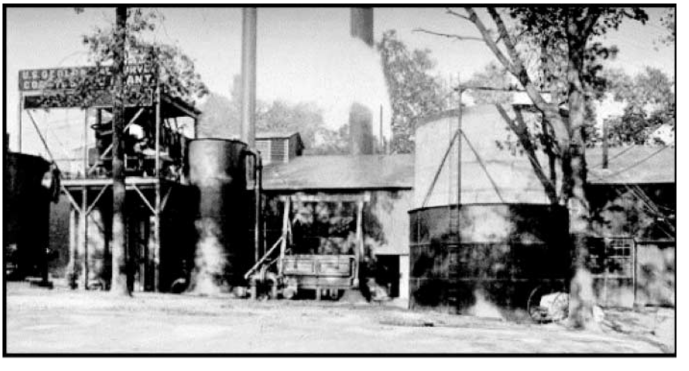

Figure 1: Town Gas Plant from 1911. Producer-Gas Plant, St. Louis, Missouri, ca 1911. Source: Fernald and Smith/US Bureau of Mines (1911).

Reforming technology development for natural gas and similar fossil fuels proceeded along two technical lines:

- steam methane reforming, an endothermic process that requires heat addition to convert a mixture of steam and natural gas to syngas at high temperature, and
- autothermal reforming (ATR), an exothermic process to convert a mixture of steam, natural gas and oxygen into syngas and excess heat.

Improvements in steam methane reforming since its introduction have been made through increasing operating pressure and temperature by development of new catalysts and reactor materials.

Combining the two steps of syngas production and separation of hydrogen and CO_2 is a well-established technology mainly used in production of syngas for ammonia production. The first system was introduced in the 1940s. It used low-pressure steam reforming followed by compression to 15 barg and CO_2 separation from the H_2 through an amine separation process using 20% monoethanolamine (MEA). In the mid-1950s, a separation technology using hot potassium carbonate was introduced and in the late 1970s, activated MDEA solvent was introduced leading to a significant improvement in energy efficiency.

The largest ammonia plants (Figure 2) produce about 2000–2200 ammonia t/day, which requires hydrogen production of about 150,000–200,000 Nm^3/h or 450–600 MW (LHV). Approximately 1,000,000 tpa of CO_2 is captured in the largest plants and compressed to 160 barg for use in urea production.

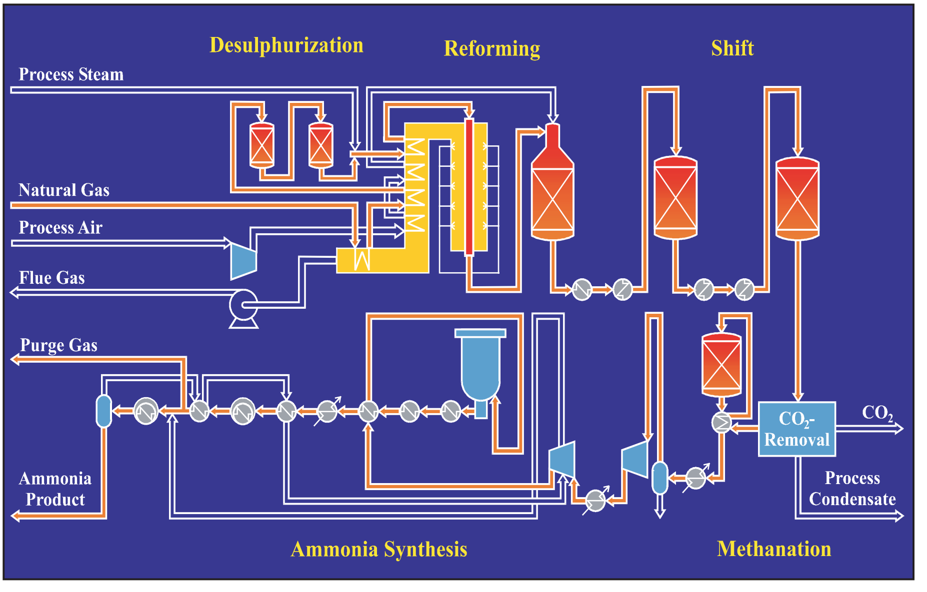

Figure 2: Ammonia process scheme. Source: Haldor Topsoe.

State-of-the-Art Pre-combustion CO_2 Capture Technology
One of the first attempts to develop a pre-combustion CO_2 capture process was in 1998, when Norsk Hydro launched the *Hydrokraft* project (Figure 3). The concept was based on air-blown ATR technology to produce a mixture of hydrogen and nitrogen for use as fuel for a large gas turbine. The project gave important insights into pre-combustion technology and into the pre-combustion base line technology.

Pre-combustion technology is a very complex process involving a number of catalytic steps, heating to high temperatures and cooling to low temperatures. Improvements that favor one part of the process might be a disadvantage for another part. As an example, reforming is favored by low pressure, but separation of CO_2 is favored by high pressure. Energy efficiency is favored by low steam addition but hydrogen production is favored by high steam addition. The energy loss is significant—almost 25% of the energy is lost in present pre-combustion processes. Further, the investment required to build a pre-combustion plant is tremendous. A combined cycle gas turbine power plant using pre-combustion processes to make hydrogen fuel will be double the cost of the same facility fueled with natural gas.

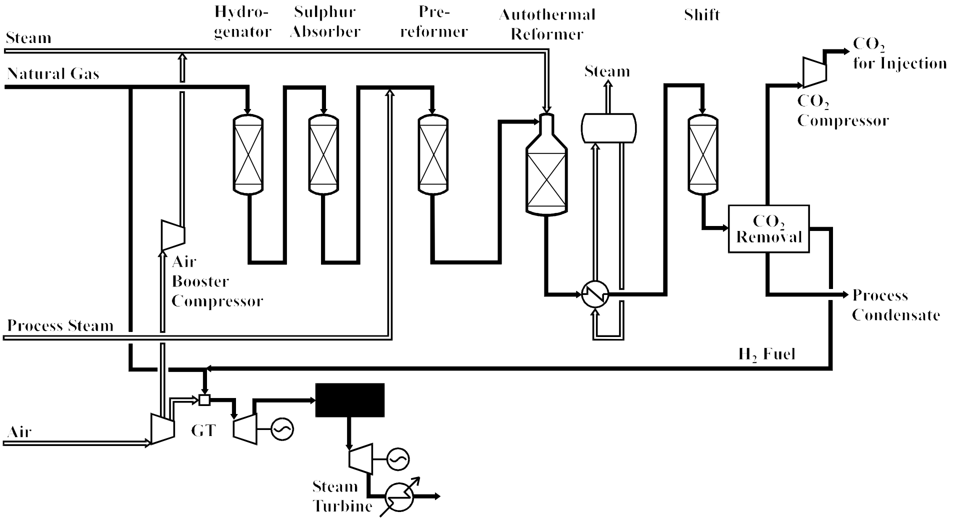

Figure 3: The “Hydrokraft” concept or Integrated Reforming Combined Cycle (IRCC)—Pre combustion baseline technology. Source: Haldor Topsoe/Hydro.

Technology Program Development

The pre-combustion team used their understanding of historical hydrogen production of other current “State-of-the-art” technology to develop two approaches for improving the pre-combustion technology:

system optimization by use of well-known technologies, and
new technologies based on advanced separation combining both reaction and separation.

Since different technologies are at different levels of development and different risk factors are associated to the technologies, a key challenge was to define a program with a balanced portfolio—taking into account that success was a result of the balance between risk and potential benefits. The team agreed to invest in technology with less potential and lower risk but with a higher chance for success.

As mentioned previously, the core technologies of pre-combustion technology have been developed for commercial markets for more than 50 years. This has created a large industry and a core area in many universities and institutes. The challenge for the team was to identify the best players in the market and to engage them in the program.

During the bidding phase, all the technology providers were requested to define a program scope that would bring the technology to a “proof-of-concept” stage. However, in order to monitor progress and differences in timing and be able to (re)-direct the program, a stage gate process was adopted. Each project identified critical milestones, e.g. material durability and performance, for different stages in the development work—called stage gate criteria. The definition of stage gate criteria was established in collaboration between the CCP and the technology suppliers. The stage gate process made decision taking and choices easier.

PRE-COMBUSTION TECHNOLOGY DEVELOPMENT PROGRAM

Based on the review and evaluation of technology and working with potential technology suppliers, the program outlined in Table 1 was defined in 2001.

TABLE 1
PRE-COMBUSTION TECHNOLOGY STUDIES STATUS: JAN 2004

References	Project title	Co funder	Technology provider	Status
1.2.1.1	Sulfur-tolerant membrane study	DoE	Fluor, SOFCo, Eltron, TDA Research, CSM, ECN, University of Cincinatti	Did not pass complete stage gate review in April ’03. Entered into phase II with reduced and revised scope. Eltron, Fluor and SOFCo were remaining technology providers. Project completed.
1.2.1.2	Hydrogen membrane reactor	EU	BP, Norsk Hydro, KTH, Sintef, University of Twente, Institute for membrane technology, University of Zaragoza	Passed stage gate review. Project completed
1.2.1.3	Hydrogen membrane reformer	Klimatek	Norsk Hydro	Passed stage gate project completed with successful “proof-of-concept” test
1.2.1.4	Precombustion membrane reactor study	CCP	Haldor Topsoe	Completed in Feb. 2001
1.2.2.1	Advanced technology for separation and capture of CO_2 from gasifier process producing electrical power, steam and hydrogen	DoE	Fluor Federal	Completed Oct 2003
1.2.3.1	Study of gas turbine retrofit requirements to burn decarbo-nised fuel (hydrogen)	DoE	General Electric	Completed Dec 2003
1.2.3.2	Standardized PCDC	Klimatak	Jacobs	Completed Dec 2003
1.2.3.3	Very large-scale autothermal reforming	CCP	Jacobs	Completed in May 2003
1.2.3.4	Advanced syngas study	CCP	Foster-Wheeler	Completed in Feb 2001
1.2.3.5	Compact reformer with advanced pressure swing adsorption system for hydrogen fuel production	DoE	Davy/APCI	Compact reformer dropped. Advanced PSA study completed Dec 2003

(*continued*)

TABLE 1
CONTINUED

References	Project title	Co funder	Technology provider	Status
1.2.4.1	Capture study integrated reports	DoE	ARI	Completed
1.2.5.1	Generation of H_2 fuels	Klimatak	IFE	Completed Feb 2002
1.2.6.1	Production of hydrogen fuel by sorbent-enhanced water gas shift reaction	DoE	Air Products and Chemicals	Passed phase II stage gate review. Phase III completed with "proof-of-concept" test

The technology development program was carried out over the 2002–2003 timeframe and is summarized here and detailed in the following chapters.

The results indicate that the technologies in the membrane area advanced much more than anticipated given the short time (16–24 months) for actual development work. The risk element was high with a reasonable likelihood of failure. Even so the membrane developers have overcome significant barriers and are well positioned to continue the work.

Sulfur-Tolerant Membrane Study (Table 1, Ref. 1.2.1.1)
The study objective was to develop a sulfur-tolerant membrane operating at water gas shift conditions. Four membrane developers were given 1 year to develop a membrane with significant flux and selectivity for a sour syngas. None of the developers reached the target; however, a promising membrane for sweet syngas condition was identified. The pre-combustion team agreed to re-direct the program and continue the development for sweet syngas application. The program continued with Eltron as the membrane developer, SOFCo as the commercial reactor designer, and Fluor as responsible for process integration.

The development of a novel low-cost compact design for a membrane water gas shift reactor and improved membrane for a water gas shift reactor with selectivity and flux was achieved. This technology will lead to reduced reactor and membrane costs in the US DOE refinery scenario and the technology shows a potential of 30–35% reduction in CO_2 avoided cost when using refinery off-gas.

The team considers this technology promising with medium potential and medium risk. They recommend that the work be continued and to, also continue searching for a sulfur-tolerant hydrogen membrane.

It should be noted that a concept based on gasification of heavy fuel oil was also developed. The CO_2-avoided cost was higher than the amine post-combustion baseline technology and was not pursued further.

Hydrogen Membrane Reactor Technology (Table 1, Ref. 1.2.1.2)
A consortium of European membrane developers was created with a common task of developing novel hydrogen membranes that could be used in pre-combustion applications—the CCP EU refinery scenario. The membrane types were ultra-thin Pd-membranes, silica-based ceramic membranes, and Pd-coated zeolites. The most promising membrane was the dense Pd/Ag membrane in which a 1 μm thick film was manufactured by a method developed by SINTEF and deposited on a porous stainless steel support tube. Significant advancement was also achieved in the silica-based ceramic membrane where a selectivity of 1000 was obtained.

A reactor concept incorporating the Pd-membrane was developed with an associated process scheme for production of a hydrogen fuel mixture for heaters and boilers. The technology demonstrates high energy and CO_2 capture efficiency and low cost. A CO_2 reduction cost of 25–30% using refinery off-gas was achieved.

The team has recommended that work on the Pd-membrane with a focus on long-term testing of stability and performance be continued.

Hydrogen Membrane Reactor Technology (Table 1, Ref. 1.2.1.3)

Norsk Hydro was selected to develop a technology based on high-temperature ceramic hydrogen membranes for combined cycle gas turbines as part of the Klimatek-funded NORCAP project. The technology principle is similar to some of the concepts being studied by the Oxyfuel team for oxygen-conducting ceramic membranes. The first phase of the project aimed at developing a membrane that could achieve significant flux in order to meet the CCP targets. This work was done in collaboration with the University of Oslo and SINTEF. At the end of Phase 1, a membrane was synthesized with sufficient flux. The membrane reformer system showed untouchable performance in the NORCAP Norwegian scenario with very high energy efficiency (approximately 90–91% LHV), low NO_x formation—5 ppm vol. and a potential CO_2-avoided cost reduction of 50–55%. Proof-of-concept tests confirmed hydrogen flux above expectations. The team recommended continued work on the technology in the extended Klimatek program for 2004.

Advanced Technology for Separation and Capture of CO_2 from Gasification, Producing Electrical Power, Steam and Hydrogen (Table 1, Ref. 1.2.2.1)

Fluor completed a complete study of pre-combustion technologies for a petroleum coke fired IGCC with production of steam, electricity and hydrogen. Uncontrolled and baseline cases were established, several pre-combustion technologies were screened, and one technology was selected for detailed design and costing. The results showed that with conventional technology, a CO_2 avoided cost of approximately $15 per ton could be obtained. This gives very little room for improvement. Screening of different pre-combustion options was based on multiple criteria, e.g. CO_2 recovery above 85%, hydrogen delivery at gas turbine pressure, sulfur tolerance, sulfur content in CO_2 stream and so on. Using these criteria, the Fluor CO_2LDSEP was seen as the most suitable option. Due to confidentiality issues, the capital cost was assessed by a sensitivity analysis—showing that the CO_2 avoided cost for the technology was in the range $10–20 per ton.

Results indicate that very low CO_2 avoided cost can be obtained in US DOE Canadian scenario by adopting pre-combustion technology—in the range $10–15 per ton CO_2.

Gas Turbine Retrofit Requirements to Burn Decarbonised Fuel (Hydrogen) (Table 1, Ref. 1.2.3.1)

One of the critical success factors for pre-combustion technology is that hydrogen-rich fuel can be used in multiple combustion processes. Hydrogen-fuel for use in gas turbines' combustors is an area that requires special attention in terms of performance, lifetime, and cost. The leading gas turbine supplier for syngas fuel combustors was selected to conduct the study—General Electric. The study results were very encouraging and, in particular, retrofit of gas turbines was confirmed to be feasible. In addition, an improved heat rate of 15% was estimated which will reduce the size of the pre-combustion plant and increase overall energy efficiency for a projected power plant. Changing from natural gas to hydrogen-rich fuel, GE estimated that a 50% NO_x reduction could be achieved and for some hydrogen fuel mixtures single-digit NO_x ppm levels can be obtained.

Standardized Pre-combustion De-carbonisation (PCDC) Technology (Table 1, Ref. 1.2.3.2)

The pre-combustion team initiated a study to evaluate cost-reducing options for pre-combustion baseline technologies. The focus was on cost savings from repeat design, modularization, mechanical codes, pre-fabrication and so on. The results were somewhat disappointing only demonstrating cost savings in the order 15–20%. Further work in this area should focus on rotating equipment, which contributed 60% to the total installed cost.

Very Large-Scale Autothermal Reforming (Table 1, Ref. 1.2.3.3)

A key feature of pre-combustion technology is the potential of designing very large capture plants in a central location with distribution of the hydrogen fuel to combustion operations thus obtaining the benefits from economy-of-scale. The study confirmed that a pre-combustion technology could be built for the Alaska scenario—capturing over 2 million tpa of CO_2 and producing more than 750 MW of fuel. The team felt, however, that the proposed process design was not optimal for the Alaska scenario and further work would be needed. Economic modeling estimates showed that the CO_2 reduction potential was less than 15%. However, looking at the option at a similar maturity as the post-combustion baseline technology—one conclusion from the work could be that pre-combustion is preferred over post-combustion technology.

Advanced Syngas Study (Table 1, Ref. 1.2.3.4)

Several technologies that are commercially available or close to commercialization were studied as the baseline technology for the Norwegian scenario—Integrated Reforming Combined Cycle technologies. The study results demonstrated limited potential whether for adopting new technology or for optimizing the concepts. However, the study showed that a pre-combustion baseline was lower cost technology than post-combustion but had higher energy consumption.

Compact Reformer with Advanced Pressure Swing Adsorption System for Hydrogen Fuel Production (Table 1, Ref. 1.2.3.5)

The scope of the work was reduced to evaluate only the advanced pressure swing adsorption system since the compact reformer developer (Davy) would not agree to the needed contract terms to allow integrated analysis. Results showed that pressure swing adsorption cycles that couple hydrogen purification with carbon dioxide recovery system offer higher hydrogen recovery with the same number of adsorbent columns. It was determined that a single-train adsorption system can provide 0.8 million tpa of CO_2 at up to 99.7% purity and with a CO_2 recovery of up to 93%. The economics of the technology and integration in a complete pre-combustion scheme is recommended.

Generation of H_2 Fuels (Table 1, Ref. 1.2.5.1)

A process scheme using CaO as an oxidant to drive the reforming reaction was developed for the Norwegian and EU refinery scenarios. The project goal was to estimate CO_2 capture and energy efficiency to be expected from the technology. Results showed that for combined cycle gas turbines, the technology could not obtain satisfactory energy efficiency reaching only 40% (LHV). For heater and boiler options, energy efficiency was estimated to be approximately 82% (LHV) if an electricity credit is included. A CO_2 capture efficiency of 90% could be obtained for both applications. CaO reacts to form $CaCO_3$ and must be converted back to CaO by calcinations for recycle to the reforming reactor. The key challenge is to develop a reactor system that can recycle solid materials efficiently. The technical risk associated with development of the technology must be considered high and fundamental studies and lab testing must be conducted before pilot testing can be realized. The recommendation from the team is to study the refinery case in more detail and establish a cost estimate for the process before considering laboratory development work.

Production of Hydrogen Fuel by Sorbent-Enhanced Water Gas Shift Reaction (Table 1, Ref. 1.2.6.1)

The leading adsorbent material ADS1-2 has a CO_2 removal capacity of up to 1.1% in PDU cyclic testing. A new adsorbent with the potential for significantly higher CO_2 capacities than other adsorbents has been identified. Further development of this adsorbent could lead to significant improvement of the sorption-enhanced water gas shift reactor scheme for gas turbine applications like the Alaska scenario or the Norwegian Scenario. The technology demonstrated significant cost reduction—in the range 30–35%—compared to the baseline technology. The technology is considered to be at a more mature stage than membrane technologies and has high potential. The team recommends continuing work on this technology.

CONCLUSIONS

Pre-combustion Technology Application to the CCP Scenarios

A key advantage of pre-combustion technology is its fuel flexibility and ability to convert all types of fossil fuels into syngas. That alone makes pre-combustion technology the only capture technology applicable to all of the CCP scenarios. Another advantage of the pre-combustion technology is that hydrogen fuel production and CO_2 capture take place in a dedicated plant at a central location yielding significant economy of scale compared to the other capture technologies. Each CCP scenario includes retrofit cases. There was concern that pre-combustion technology could not be retrofit to gas turbines. Our studies show that turbines can be retrofit to burn hydrogen fuels (GE study).

Three of the most promising technologies were applied to CCP scenarios using technology provider information:

- Hydrogen Membrane Reformer for NorCap scenario.
- Membrane water gas shift reactor for the UK refinery.
- Sorption-enhanced water gas shift reactor for the Alaska scenario.

Fluor evaluated the technologies in the subject scenarios. Their studies included: the integrated design, quality assurance, and cost estimation. The study created a unique platform for comparison against the baseline technologies thus giving new insights on how the technologies performed in the given scenario and credibility to the cost reduction potential estimated by the CEM team.

Economics

Economic modeling results discussed in Chapter 4 of this Volume showed that three novel pre-combustion technologies had a significant potential to reduce CO_2 avoided cost compared to the baseline technology. The Hydrogen Membrane Reformer, assessed in the NorCap scenario, demonstrated a CO_2 avoided cost reduction potential of 60% compared to the baseline technology. This is equal to an annual saving of approximately \$35 million for a 400 MW power plant.

The membrane water gas shift system was assessed in the Canadian gasification scenario for two different fuels (refinery fuel oil and refinery off-gas). The results were remarkable. In the refinery fuel oil case, the CO_2 avoided cost increased when compared to the baseline technology because both energy loss and capital cost required to gasify the fuel oil was much higher than anticipated. In the refinery off-gas case, the much more efficient reforming process, ATR, could be used. For that case, the CO_2 avoided cost was reduced by 30–40% compared to the baseline technology.

Sorption-enhanced water gas shift was assessed in the NorCap and Alaska scenarios. In the NorCap scenario up to 44% cost reduction was achieved by using air-blown ATR, but for the Alaska scenario only 19% cost reduction was achieved. One important remark is that the improved gas turbine efficiency gained by switching from natural gas to a hydrogen/nitrogen fuel-mix was not taken into account in the Alaska scenario. Further, the Alaska scenario requires a very special design due to the extreme climatic conditions and location—this issue reduces the relative improvement when using new technologies.

Commercial Value—Present and Future

Pre-combustion technology for CO_2 capture accommodates a broader potential than the other capture technologies. The technology is widely applicable in any syngas production process such as: methanol, synfuel, ammonia, and hydrogen. Thus technology improvements made by the CCP can be adopted as well in these areas. As an example, a large GTL plant costs about \$1 billion and 60% of the cost relates to the syngas technology.

Significant improvement in hydrogen production technologies could be the base for low-cost hydrogen for future fuel cell vehicles. Hydrogen production with capture and storage of CO_2 will “bridge-the-gap” towards the renewable hydrogen economy.

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