Technology Options and Integration Concepts for Implementing CO_2 Capture in Oil-Sands Operations

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Summary

The majority of the technology development for CO_2 capture and storage (CCS) is driven by the electric-utility industry, in which the emphasis is on large centralized units for electric-power generation with coal as the primary fuel. The implementation of CCS in oil-sands operations has significant potential to provide meaningful carbon-emissions reductions. This paper presents various concepts for integrating leading CO_2-capture techniques to bitumen-extraction and -upgrading processes. The main carbon-capture technologies are reviewed, and their relative advantages and disadvantages for implementation in bitumen mining, thermal bitumen extraction, and bitumen upgrading are discussed, leading to a qualitative assessment of their suitability for each oil-sands process.

Introduction

This paper is intended as an in-depth briefing on carbon capture for non-CO_2-capture experts possessing reasonable knowledge of oil-sands operations. It is qualitative in nature and focuses on capturing CO_2 from stationary processes found in oil-sands operations. Our objectives are three-fold: (a) to provide the reader with an overview of the landscape of carbon-capture technology, (b) to highlight the opportunities and challenges of implementing CO_2 capture in oil-sands operations, and (c) to provide our perspective on the comparative suitability of existing capture technologies as carbon-mitigation solutions for oil sands.

The implementation of CCS in oil sands is a subject demanding scientific and technical scrutiny. Few studies have examined the subject of carbon emissions from oil-sands operations (Ordorica-Garcia et al. 2007; Charpentier et al. 2009). CO_2-capture-implementation issues in oil sands at the process level are insufficiently covered in the existing literature, while the majority of CCS papers focus on power generation and not on oil-sands applications.

The study covers the leading carbon-capture technologies: precombustion, post-combustion, oxyfuel combustion, and chemical looping combustion. The first part provides an in-depth analysis of each of these technologies, covering their fundamentals, status, and outlook, drawing from several sources. The second part discusses integration options of the four capture technologies with the three main oil-sands processes: mining, steam-assisted gravity drainage (SAGD), and upgrading. The paper presents an evaluation of capture solutions for each process according to technical suitability and highlights the advantages and challenges of various integration concepts.

Oil-Sands Operations Overview

Canada's oil-sands operations have experienced substantial growth in the last decade. Going forward, it is likely that this trend will continue. Because of their energy-intensive nature, increased bitumen-extraction and -upgrading operations may lead to record fossil-energy demands and greenhouse-gas emissions.

In parallel, domestic and foreign environmental concerns have led the provincial government and industry to look for solutions to mitigate the carbon emissions of oil-sands operations. CCS technology has recently emerged as the leading option. At the time of writing, four large CCS industry projects are candidates to receive governmental support totalling USD 2 billion (Scotton 2008).

This study deals with the integration of carbon-capture technologies with three fundamental oil-sands processes: bitumen mining (including extraction through the hot-water process), thermal extraction through SAGD, and bitumen upgrading to synthetic crude oil (SCO).

Bitumen Mining. Bitumen mining involves digging the bitumen-bearing sand from the ground, using shovels and trucks to move it. The sand is then slurried with warm water and often transported through pipeline to an extraction plant, where the sand is treated with hot water, steam, and chemicals to yield raw bitumen. The bitumen is then diluted with light hydrocarbon fractions and pipelined to market or to upgrading into SCO.

The process uses large amounts of hot water, and modest amounts of steam and electricity. These energy commodities typically come from cogeneration plants and steam boilers, which are most often fuelled by natural gas. The CO_2 emissions in mining come from mobile and stationary sources. Mobile sources include trucks and shovels, which are not amenable for carbon capture. The key characteristics of the flue gas from stationary sources in mining operations are large volumes of warm, low-pressure gas, with low CO_2 concentrations (approximately 5 mol%, dry basis).

Previous work (Ordorica-Garcia 2007) has estimated that on average, CO_2 emissions related to hot-water production account for 35%, electricity is responsible for 29%, and steam-related emissions are only 13% of the total carbon emissions in mining operations. The remaining 23% corresponds to emissions from mobile sources. The analysis presented here is limited to the implementation of carbon-capture technology in stationary sources, hence the mitigation of mobile or fugitive sources of CO_2 is not covered.

SAGD Extraction. Although there are various in-situ methods for extracting bitumen from underground reservoirs, the dominant technology is SAGD. SAGD extraction involves drilling horizontal well pairs in the oil-sands reservoir. Steam injected through the upper well rises through the deposit and heats the bitumen. The hot bitumen separates from the sand and flows along with condensed steam into the lower well and is then pumped to the surface. Bitumen is separated from other fluids removed from the reservoir and diluted with light oil fractions to facilitate its transport.

Most commercial SAGD extraction processes produce steam in once-through gas-fired steam generators. Cogeneration plants also can be used to supply power and steam to the process. The energy demands of SAGD processes consist overwhelmingly of steam, with electricity usually accounting for less than 5% of the

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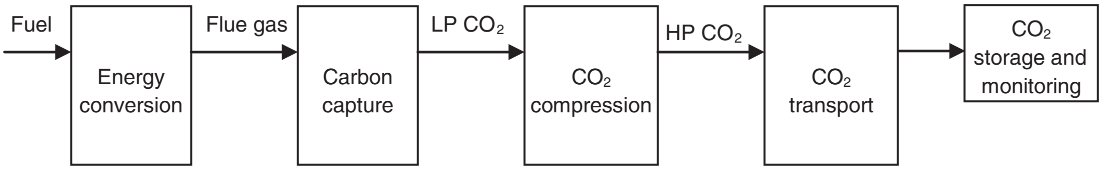

Fig. 1—CCS system components.

total. The CO_2 emissions from SAGD bitumen extraction are largely set by the amount of steam injected per barrel of bitumen produced. This parameter, known as the steam/oil ratio (SOR), typically ranges from 2 to 5. The higher the SOR, the higher the CO_2 emissions. The emissions distribution of a typical SAGD operation is 97% from steam production and 3% caused by electricity generation (Ordorica-Garcia 2007). The flue gas produced has a typical CO_2 concentration of approximately 5 mol% (dry basis).

Bitumen Upgrading. Commercial bitumen-upgrading operations rely on thermal or/and hydrogen cracking methods. Thermal methods use coking, a carbon-rejection process, whereas hydrocracking processes involve hydrogen addition, to yield lighter oil fractions. Before cracking, the diluted bitumen undergoes distillation to recover the diluent, which is often recycled. After coking or hydrocracking, the resulting oil fractions are hydrotreated to remove sulphur and other contaminants. These fractions are blended into synthetic crude.

Upgrading is the most complex and energy-intensive of the oil-sands operations. The process operations involved and their energy demands are varied. For instance, distillation units use steam and/or process fuel; cokers require combinations of steam, electricity, and process fuel; while hydrotreaters and hydrocrackers require hydrogen, electricity, and process fuel. These energy commodities are commonly produced in cogeneration plants (electricity and steam), steam methane reformers (hydrogen), or boilers (steam). Additionally, process fuel is burned in a variety of smaller, distributed units (e.g., furnaces and heaters). The fuel is overwhelmingly natural gas, although internally produced fuel gas is also burned in upgraders.

Concerning CO_2, H_2 production is responsible for more than 50% of the total emissions from upgrading, while steam generation accounts for approximately 40%. Electricity generation and process-fuel burning contribute less than 10% to the total carbon emissions of a coking-based upgrader. In hydrocracking upgrading operations, hydrogen accounts for more than 67% of emissions, followed by steam, which represents approximately 20% of the total, with the balance consisting of power generation and process-fuel burning (Ordorica-Garcia 2007).

Hydrogen production is the main source of carbon emissions in bitumen-upgrading operations. For this reason, the study includes the option of gasifying upgrading residues (e.g., pitch and petcoke) for hydrogen production. Currently, methane reforming is the dominant technology, but future upgrading projects may use gasification instead. From a CCS perspective, the choice of hydrogen-production technology impacts the volume and quality of the CO_2 generated. Gasification would yield larger amounts of CO_2 at higher concentrations than steam methane reforming (SMR). The CO_2 content of the former ranges from 36 to 41%, whereas that of the latter is less than 30 mol% (dry basis). In contrast, other CO_2-bearing streams in upgrading (e.g., gas-fired boilers, furnaces, and cogeneration plants) feature CO_2 concentrations of 5 to 8 mol% (dry basis) (Ordorica-Garcia et al. 2009).

Technologies for CO_2 Capture

There are four main approaches to capture CO_2 from fossil-fuel-fired energy systems: precombustion, post-combustion, oxyfuel, and chemical looping combustion. These technologies are suitable for large, stationary carbon emissions point sources (e.g., fossil-fuel power plants and hydrogen plants). However, capture is only one link in the CCS chain. A CCS system also requires CO_2 compression, a means to transport the CO_2, and a storage site, as illustrated in **Fig. 1.** While the paper focuses only on the capture component, the complexity and effort required to remove CO_2 from the atmosphere successfully go beyond carbon capture.

Precombustion. In precombustion capture, CO_2 is separated from a gaseous fuel mixture under reducing conditions before its combustion. Most commercially available technologies rely on liquid solvents for CO_2 absorption, usually by scrubbing a gas stream with physical solvents (Kohl and Nielsen 1997). A simplified diagram of the process is depicted in **Fig. 2.** Post-combustion capture is often preceded by the conversion of a carbonaceous fuel (e.g., natural gas, coal, biomass, or oil) to a gaseous mixture consisting primarily of H_2 and CO, called syngas. The fuel-conversion step during gasification or reforming is endothermic and requires supplementary heating, typically supplied by partial oxidation of the fuel. In CO_2-capture applications, high-purity O_2 obtained from an air-separation unit (ASU) is often used as oxidant, yielding the high temperatures required to produce syngas.

The CO in the syngas is converted to H_2 and CO_2 through the water/gas shift (WGS) reaction. The WGS reaction is facilitated by commercially available catalysts, suitable for use with low and high sulphur (H_2S and COS) concentrations. The CO_2 is separated through physical absorption with a solvent. The CO_2-rich solvent is regenerated by pressure reduction, releasing the CO_2, and the regenerated solvent is reused for CO_2 absorption. The recovered CO_2 is dried and compressed to facilitate its transport and storage, while the hydrogen-rich stream can be combusted for electricity generation or used for synthesis of chemicals.

Syngas production from gasification and partial oxidation is generally carried out at high pressure, resulting in high CO_2 partial pressures. For this reason, physical solvents are the favoured means to capture CO_2 in precombustion applications. Physical absorption is subject to Henry's law, which results in a linear dependency between the CO_2 partial pressure and the solvent capture effectiveness.

The Rectisol® process, invented by Lurgi in the 1950s, is one of the most commercially used for H_2S and CO_2 removal (Weiss 1988). It uses methanol at low temperatures, (−20 to −60°C). The low temperatures favour increased solubility of the acid gases (CO_2, COS, and H_2S) present in syngas. Most of the cooling is attained by methanol expansion during flash regeneration, but additional refrigeration is needed. Operation at higher temperatures would result in considerable solvent losses because of its high vapour pressure. For locations where these low temperatures are readily achievable during winter, Rectisol would have an important advantage over other solvents for CO_2 capture.

Other common precombustion-capture processes employ organic solvents with high molecular mass and boiling points (Hiller et al. 2000). The most prominent processes are: Selexol^{TM}, using a mixture of di-methyl ethers of polyethylene glycol (licensed by UOP); Purisol, using N-methyl-2-pyrrolidone (licensed by Lurgi); and the Fluor Solvent using propylene carbonate (licensed by Fluor Daniel). These processes are operated at near-ambient temperatures because low temperatures would increase the solvent viscosity significantly. CO_2 solubility at ambient temperatures is comparable for all these solvents.

Status. All the technologies related to precombustion capture (i.e., coal gasification or natural-gas reforming, WGS reaction, CO_2 separation, and compression) have been proved at a

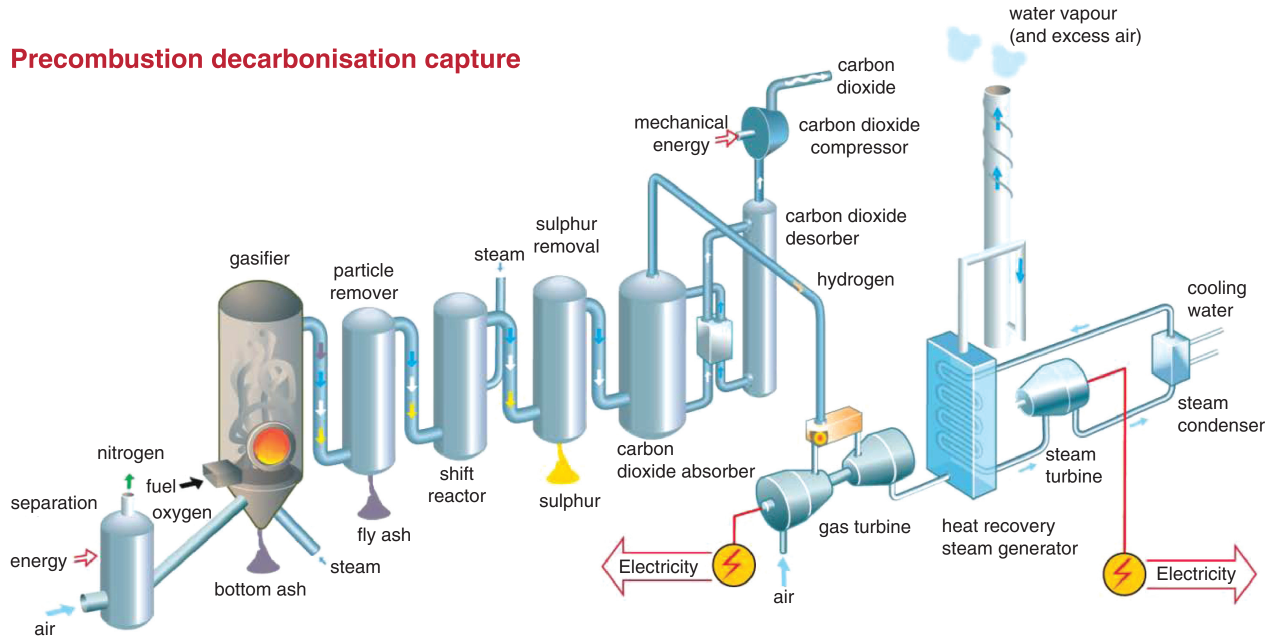

Fig. 2—Precombustion CO_2-capture schematic (Strömberg 2005).

commercial scale in multiple sites around the world. CO_2 is captured predominantly to meet chemical synthesis or product specifications or for enhanced oil recovery (EOR). However, no commercial plant has been built yet specifically to capture and store CO_2 to reduce greenhouse-gas emissions. In this section, we review a selection of key operational industrial plants and scheduled pilot/demonstration precombustion projects.

The Great Plains Synfuel plant near Beulah, North Dakota, USA, uses lignite as feedstock for the synthesis of substitute natural gas (Honea 1985) and has been in operation since 1984. It features Lurgi dry-ash gasifiers, a sour WGS conversion, and Rectisol for CO_2 separation (Kohl and Nielsen 1997). It produces 3.3 million t of CO_2 annually, of which 1.0 to 1.5 million t are compressed, pipelined, and injected in the Weyburn oil field in Saskatchewan for EOR.

Since 1998, at its Pernis refinery in the Netherlands, Shell converts heavy refinery residues into hydrogen for hydrocracking and electricity using dry-fed Shell gasifiers (Zuideveld et al. 1998). The Rectisol process is used for CO_2 recovery; approximately 150,000 t/a are used in industrial processes and since 2005, part of it is transported to nearby greenhouses for CO_2 fertilization (380,000 t/a, mainly in the summer). The increased CO_2 concentration in greenhouses was previously attained by combustion of natural gas.

Farmland Industries in Coffeyville, Kansas, USA, in operation since 2000, uses petroleum coke as feedstock for the synthesis of ammonia and urea-ammonium-nitrate (Ferguson et al. 1999). The petcoke is converted into syngas in a water slurry-fed General Electric (formerly Texaco) gasifier. The plant uses the refurbished gasifier section of the cool water-integrated gasification-combined cycle (IGCC) demonstration plant and uses Selexol to separate CO_2 from the syngas. Some of the CO_2 is used to produce urea-ammonium-nitrate, and the remaining CO_2 is vented to the atmosphere.

In addition to commercial plants, pilot and demonstration precombustion-capture plants totalling approximately 800 MW are in operation around the world. **Table 1** provides a selection of the most recent projects.

Future Prospects. All technologies related to precombustion CO_2 capture are currently employed on a large industrial-scale (i.e., WGS reactors, CO_2 scrubbing with physical solvents, CO_2 compressors, and hydrogen-rich syngas combustion in gas turbines). The main incentives to separate CO_2 at this scale are the production of hydrogen from a carbon-rich feedstock or the adjustment of the H_2/CO ratio in synthesis processes (e.g., chemical or fuel production). In the latter, near-pure CO_2 is often vented to the atmosphere. This suggests that precombustion capture is more likely to be deployed for synthesis of chemicals or fuels because the incremental implementation costs are significantly lower than for electricity generation.

The use of precombustion CO_2-capture technologies in electricity generation is currently limited to pilot-scale CO_2-capture plants. The most notable prospective commercial plants featuring precombustion capture and CO_2 storage are

- The Hydrogen Power Project in Abu Dhabi is a joint-venture between Masdar and BP and involves a 420-MW_e hydrogen-fired-combined cycle power plant (Hill 2009). Hydrogen is obtained by the reforming of natural gas; approximately 1.7 million t/a of CO_2 will be used for EOR. The commissioning of the plant has been postponed and is foreseen in 2016 or 2017.
- The HECA project in southern California was an initiative of BP and Rio Tinto, which will be continued by SCS Energy. This IGCC power plant will use a petcoke/western-bituminous-coal blend (25 to 100% petcoke) as fuel (SCS Energy 2009). The net electrical output is approximately 250 MW_e; an additional 100 MW_e that is natural-gas generated will be available for peaking. The CO_2-capture rate is 90%; more than 2 million t/a will be used

TABLE 1—REPRESENTATIVE PRECOMBUSTION CO_2-CAPTURE PILOT AND DEMONSTRATION PROJECTS

Project	Country	Size (MW_e)	Status	Gasifier	Fuel	CO_2 Capture Details
Elcogas (García-Peña and Coca 2009)	Spain	283	Started operations in 2009	Prenflo	Coal/Petcoke	36.5 ktonnes CO_2/y
NUON (de Kler 2009)	Netherlands	253	Started operations in 2010	Shell	Coal/biomass	10 ktonnes CO_2/y
GreenGen (Xu 2008)	China	250	Started operations in 2010	TPRI	Coal	97 ktonnes CO_2/y

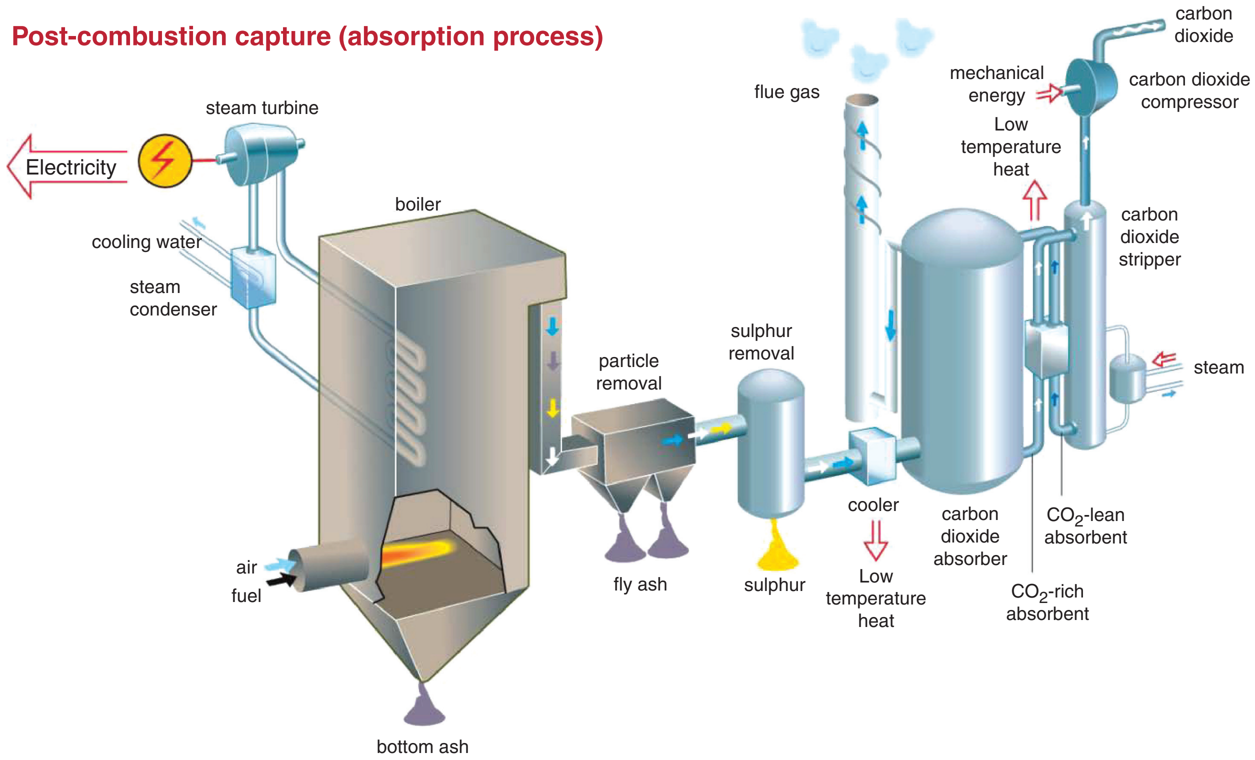

Fig. 3—Post-combustion-capture schematic (Strömberg 2005).

for EOR in a nearby oil field. The first operation is scheduled for 2015.

- The Zerogen project in Australia plans to build a commercial-scale IGCC with a net output of 530 MW_e (Tarr 2009). The plant will capture between 65 and 90% CO_2 using Selexol and is scheduled to be completed by 2015.

The plans for commercial-scale precombustion-capture power projects are subject to great volatility because of the large capital expenses involved and constrained financing conditions at the time of writing.

Research and development of advanced precombustion CO_2-capture technologies are dedicated mainly to electricity-generation applications, where the main intent is to reduce the loss in thermal efficiency caused by the energy required for capture and the associated costs. Research is focused on process intensification (i.e., the use of separation-enhanced reforming/WGS reactors and the use of membranes for low-cost oxygen production). These concepts are likely to reduce the parasitic electricity consumption that is associated with CO_2 separation and with O_2 production.

Post-Combustion. Post-combustion capture is the removal of CO_2 from flue gases downstream of emission sources. In broad terms, CO_2 can be captured using (a) liquid or solid absorption, (b) solid adsorption, and (c) membranes. The most mature post-combustion capture techniques involve liquid absorption using chemical solvents. Solid-based absorption and adsorption are in the early stages of development. Membrane technology is currently limited in application, but the research and development efforts in this area are ongoing (Bernardo et al. 2009). The discussion here is limited to liquid-absorption post-combustion capture technology.

In atmospheric air-fired combustion processes, CO_2 is often captured by means of a chemical solvent, required because of the low CO_2 partial pressures involved. Liquid-based CO_2 absorption processes operate as shown in **Fig. 3:** the flue gas is cooled and channelled to an absorber, where it flows counter current of a chemical solvent. Most of the CO_2 is absorbed within the solvent, which is then sent to a stripper, where CO_2 is recovered by application of heat, regenerating the solvent. The regenerated solvent is cooled and recycled to the absorber. The recovered CO_2 is then dehydrated and compressed for transport.

Key parameters affecting the technoeconomics of liquid post-combustion CO_2 capture include flue-gas- and solvent-flow rates, CO_2-removal rates, and energy requirements. The flue-gas-flow rate determines the absorber size and largely sets the capital costs. CO_2-removal rates affect costs and energy demands; higher rates require taller absorbers and incur larger energy demands. The former is set by the activation energy and reaction kinetics of solvents, while the latter is related to increased flue-gas, and solvent-flow rates.

Although there are numerous solvents capable of absorbing CO_2, only a few of them are adequate for post-combustion CO_2 capture. Aqueous amines and blends of amines are currently the most suitable solvents, but novel solvents are under development. The ensuing discussion will provide an overview of both.

Status. The most commercially mature post-combustion technology is Fluor's Econamine FG Plus process. It uses a corrosion-inhibited 30 wt% monoethanol amine (MEA) solution that can achieve CO_2 recoveries ranging from 85 to 95% and produce CO_2 with purities in excess of 99.95%. The flue-gas feed to the process must have low SO_x or H_2S content. The process is designed to operate at atmospheric pressure, but pressurised gas streams may be used under certain conditions (Chapel and Mariz 1999).

Another process, developed by BASF, uses activated methyldiethanolamine (MDEA) for H_2S and CO_2 removal. It is used mainly in natural-gas-processing, ammonia, and synthesis gas applications. Its advantages include low toxicity, low volatility and corrosiveness, and lower capital and operating costs because of increased CO_2 loading. BASF has developed an improved solvent that was comprehensively tested in Denmark at the Elsam power plant, within the European Union's CO_2 From Capture to Storage (CASTOR) project (IEA GHG 2008).

Mitsubishi Heavy Industries' KS-1^{TM} solvent (a proprietary hindered amine) features higher CO_2 loading capacity and lower solvent-regeneration energy requirements than MEA and is oxygen-resistant. KS-1 has lower solvent-flow rates, which translate into lower capital and operating costs than those of MEA-based processes (Yagi et al. 2006). An improved design that is said to have CO_2-capture costs 15% lower than the current process started operations in Abu Dhabi in 2009 (Kishimoto et al. 2009).

All of the previously described technologies are commercially mature and have been deployed extensively during the last 30

TABLE 2—REPRESENTATIVE POST-COMBUSTION CO_2-CAPTURE PILOT AND DEMONSTRATION POWER PLANTS

Project	Country	Size (MW_e)	Status	Solvent	Fuel	CO_2 Capture Details
RWE Power Niederaussem (Mosera et.al. 2010)	Germany	Unspecified	Began operations in 2009	MDEA	Lignite	2.6 ktonnes CO_2/y
AEP Montaineer (Vauhini 2010)	USA	30	Began operations in 2009	Chilled ammonia	Coal	100 ktonnes CO_2/y
Alabama Power Plant Barry (Ivie and Irvin 2010)	USA	25	Startup in Q1, 2011	KS-1	Coal	100–150 ktonnes CO_2/y

years. The MDEA process is in use in more than 200 plants around the world, but its main application is in natural-gas processing. The Econamine FG process is used in a wide variety of industries and has the most current operating plants of all three solvents because of its older inception date. Mitsubishi aims to have KS-1 carbon mitigation projects in power-plant applications, in addition to existing commercial plants. Current research and development programs, which are described next, focus on developing new solvents, processes, and combinations thereof. Their main goals are reducing capital costs and attaining a 50% decrease in energy demands with respect to the MEA process. **Table 2** summarizes recent pilot and demonstration projects. All the projects use coal or lignite as fuel for power production. Natural-gas power plants are not the primary target for post-combustion capture technology, but rather coal-fired plants.

Future Prospects. Although MEA is the best-known solvent, its high energy requirements (~4 GJ/t CO_2) and degradation rates are major drawbacks. To counter this, research on additives (or activators) is ongoing. Piperazine ($C_4H_{10}N_2$) has the potential to yield faster CO_2-absorption rates, lower flow rates and energy requirements, and smaller equipment. In laboratory experiments, a blend of MEA and piperazine has achieved 45% greater CO_2 capacity and faster reaction rates than conventional MEA (Dugas and Rochelle 2009).

In the short term, amine blends [e.g., MDEA-MEA and MDEA-DEA (Diethanolamine)] may achieve solvent-regeneration cost reductions of up to 67% with respect to MEA processes (IEA GHG 2008). The University of Regina leads the development of blended amine solvents for post-combustion capture. These novel solvent blends feature process designs almost identical to those of conventional MEA-based CO_2-capture plants, offering the potential to reduce commercial-deployment times.

Emerging technologies (e.g., Alstom's chilled ammonia process) are also being developed. It uses a slurry of ammonium carbonate and bicarbonate to capture CO_2 from cooled flue gas. Cooling condenses water and eliminates some contaminants, while reducing the flue-gas volume and thus increasing its CO_2 concentration. CO_2-capture levels in excess of 90% are claimed (Telikapalli et al. 2010). The process uses a low-cost reagent, unlike some amine-based solvents. The process has a low heat of reaction, high CO_2 capacity, and is tolerant to oxygen and other impurities. It does not require extremely low sulphur levels in the flue gas. CO_2 is recovered at high pressure (>20 bar), lowering compression costs. Water wash is required to scrub ammonia entrained in gas streams leaving the process, thus water requirements may be an issue in some locations.

Liquid absorption technology could also be used for multipollutant capture. For instance, a new CO_2-capture solvent developed by Cansolv (now owned by Shell) can cocapture SO_2 and CO_2 in one step, although this had not been proved at the time of writing. The solvent purportedly has lower operating costs than those of MEA. Research and development efforts intend to develop new solvents for combined NO*x*, Hg, and CO_2 absorption, ideal for coal-fired power-plant flue-gas cleanup.

Oxyfuel Combustion. In oxyfuel combustion, pure oxygen is used as the oxidant instead of air. Thereby, the resulting flue-gas stream consists mainly of carbon dioxide and water vapour. The latter can be removed by condensation; therefore, CO_2 is recovered at high concentrations. **Fig. 4** shows a diagram of a generic coal oxyfuel combustion process. Compared with air-fired combustion, the additional equipment required is a flue-gas cooler (for water condensation/removal) and an ASU for oxygen production. A certain portion of flue gas is recycled to moderate flame temperatures in the boiler, and can be extracted at various points between particle removal, water removal, and desulphurization. This involves a trade-off between obtaining a cleaner flue-gas-recycle (FGR) stream and requiring larger pollutant-control systems. The advantage of the former is that pollutant accumulation and corrosion in the boiler are reduced. In the latter case, larger gas-cleanup equipment increases capital costs. As with air-fired combustion, two main technologies for solids-fuel/oxyfuel combustion are used in utility-scale power generation: pulverized fuel boilers and fluidized bed boilers (FBBs).

In oxyfuel, the presence of nitrogen from air is largely avoided during combustion; therefore, the volume of flue gas produced decreases considerably. This yields a reduction in both flue-gas treatment-equipment size and heat losses. CO_2 concentrations of up to 92% have been recorded in pilot-scale experiments. Higher purities are difficult to reach because of air leakage into the boiler (Santos 2009).

The composition, density, and thermal capacity of oxyfuel flue gas are different from those of flue gas from air combustion. This causes substantial changes in heat transfer, flame temperature/stability, combustion efficiency, and production of pollutants within the boiler. These issues, together with the high energy requirements of the ASU plant, are the main current areas of development in oxyfuel combustion.

Status. Regarding oxyfuel boiler design, two key aspects differ greatly from air-combustion processes: changes in fuel-combustion behaviour and heat-transfer characteristics. Fuel-combustion reactivity has been studied extensively (Murphy and Shaddix 2006; Borrego and Alvarez 2007; Arias et al. 2008; Shaddix and Molina 2009) to better understand the effect of O_2 in CO_2 atmospheres on devolatilization and combustion rates. Combustion has also been modelled and validated in pilot-scale oxyfuel facilities [e.g., the CANMET vertical combustor (Chui et al. 2003) or the circulating fluidized bed (CFB) at Czestochowa University]. These studies show that the optimum O_2 concentration ranges from 30 to 35% (dry basis, depending on the fuel moisture and ash content and the FGR ratio).

Radiation is the dominant heat-transfer mechanism in pulverized coal oxyboilers. CO_2 and H_2O are responsible for most of the radiation exchange. An FGR ratio of approximately 3:1 is needed to attain a heat-transfer profile similar to that of an air-fired combustion system. In the case of FBB, this rate could be smaller, thanks to better peak-temperature control. Changes in oxyfuel boiler water walls design and optimization are still needed (Jäntti et al. 2007). Further research is under way to understand the effect of recycling flue gas better from the different possible points of the flue-gas circuit (Kather 2007).

CO_2 from oxyfuel combustion has a higher concentration of noncondensable species (e.g., O_2 and N_2) than that derived from other capture technologies (Seevam et al. 2008). Special care must be taken with NO_x, SO_2, and O_2. NO_X and SO_2 can react in the compressors in the presence of water and oxygen, forming H_2SO_4 and HNO_3. Concerning sulphur, some researchers (Croiset and Thambimuthu 2001; Kiga et al. 1997) have observed lower SO_2 formation compared with air firing of coal. However, this is highly dependent on coal composition and the FGR configuration.

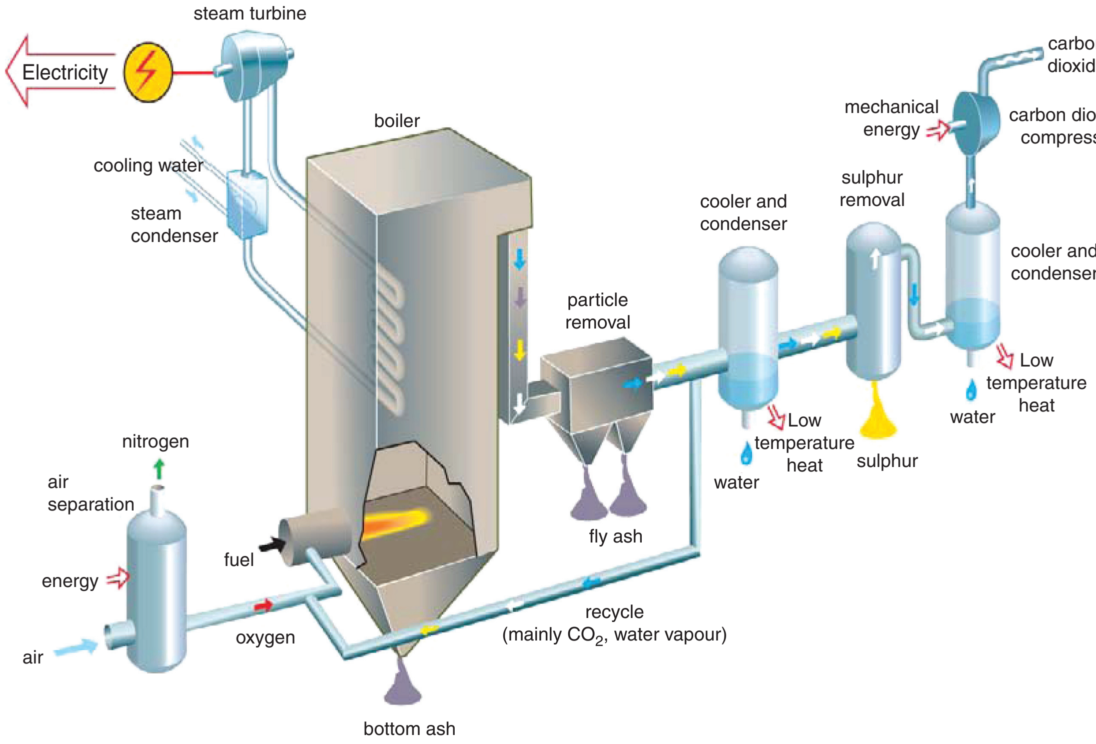

Fig. 4—Oxyfuel-combustion-capture schematic (Strömberg 2005).

Recent results show an increase in SO_3 formation and in sulphur retention in ash (Santos 2009).

Unintended air ingress into the system is a major source of impurities and the focus of research and development efforts. An experimental plant was built to study the liquefaction of CO_2 with different impurities at the Technical University of Hamburg (Köpke et al. 2009). At CANMET in Canada, a CO_2-compression train was integrated with an oxyfuel vertical combustion unit yielding 95% pure CO_2 (Zanganeh et al. 2009). The removal of noncondensable gases can reduce compression work but makes water condensation more difficult (Li et al. 2009). Leakage could also be reduced by boiler pressurization.

Upscaling oxyfuel combustion to industrial size depends heavily on the economics of large-scale oxygen production. There are various commercial technologies for O_2 separation from air, but currently, only the cryogenic method is feasible for utility-scale oxyfuel applications. The largest existing single-train ASU produces 4000 t/d, and design studies have been performed for 7000 t/d (Anheden et al. 2005).

Currently, several oxyfuel plants are planned and some of them are already operational. The most-representative pilot plants are shown in **Table 3.**

Future Prospects. Improved oxyfuel burner designs are needed to address issues including higher oxygen-feed concentration, fuel conveying, burner aerodynamics, and flame staging (Becher 2008; Lee et al. 2009). Pilot plants run by Doosan Babcock in the UK and Babcock and Wilcox in the USA, (40 and 30 MW_{th}, respectively) intend to test burner performance, turndown, startup, and shutdown, with transitions between air and oxyfuel firing (Wall 2009).

FBBs are a leading area of development (Jia et al. 2007). They feature better temperature control because of the high thermal capacity of the bed matter. Moreover, fluidized designs offer the possibility of controlling in-furnace SO_x formations with limestone, which is another important avenue of research to reduce oxyfuel sulphur-removal cost (Eddings et al. 2009; Czakiert et al. 2009; Romeo et al. 2011). The first such industrial-sized plant is a 30-MW_t boiler currently under construction in Spain. Results from this plant will form the design and construction basis for a 300-MWe oxyfuel fluidized bed plant, including CO_2 transport and storage (Kuivalainen et al. 2010). Other demonstration projects under development include: a 78-MWe boiler in Jamestown, NY, USA; a 100-MWe PC boiler (Babcock and Wilcox); a 250-MWe boiler (Vattenfall); and a 100-MWe oxyfuel PC boiler (Kepco).

Uncertain purity requirements for CO_2 geological storage are an issue for oxyfuel development, although some preliminary guidelines were obtained from the ENCAP project (Sarofim 2007). Entrained O_2 in oxyfuel flue gas is an issue for injection in depleted oil and gas fields because of the likelihood of combustion. Thus, O_2 must be removed before injection. Leading oxygen-removal research focuses on using a fuel-rich or a catalytic combustor to consume the oxygen in the CO_2 before inerts removal. Also, a distillation process could yield CO_2-stream purities of 10 ppm of O_2 (White et al. 2006).

TABLE 3—REPRESENTATIVE OXYFUEL-COMBUSTION CO_2-CAPTURE PILOT AND DEMONSTRATION PROJECTS

Project	Country	Size (MW_{th})	Status	Boiler Type	Fuel	CO_2 Capture Details
Vattenfall (Burchhardt 2009)	Germany	30	Began operations in 2008	Pilot PC	Lignite, coal	75 ktonnes CO_2/y. Storage in gas field
Total, Lacq (Aimard et al. 2009)	France	30	Began operations in 2010	Industrial. Steam for utilities	Natural gas and liquid fuels	60 ktonnes CO_2/y. Storage in depleted gas reservoir
Callide (Spero and Montagner 2009)	Australia	90	Startup in 2011	30 MWe Retrofit PC	Coal	30 ktonnes CO_2/y. Road transport to a depleted gas field

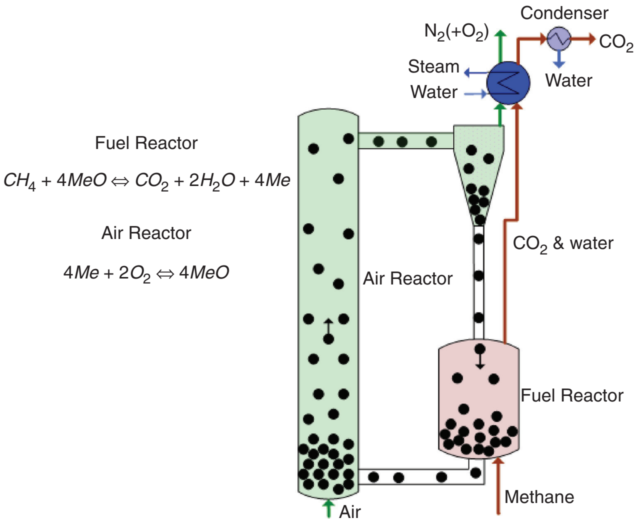

Fig. 5—Fluidized bed concept for CLC.

One of the main drawbacks of oxyfuel, compared with other CO_2-capture technologies, is the added energy for and cost of oxygen production. State-of-the-art technology based on cryogenic air separation for producing 95% purity O_2 requires approximately 200 kW-h/t O_2 (Sarofim 2007). Oxygen production thus results in energy losses equal to approximately 7 to 10% of the efficiency of a typical coal power plant. Because of its advanced maturity level, the potential of achieving higher cryogenic O_2-production efficiency is limited. To address the limitations of cryogenic oxygen production, different concepts are being investigated. Ion transport membranes, under development by Air Products (White et al. 2009), and oxygen transport membrane, patented by Praxair, have the advantage of fully avoiding N_2 from air with lower power demands. To date, however, membrane technology for oxygen production is in an early stage of development and is currently limited to small-scale (pilot) applications.

Chemical Looping Combustion (CLC). CLC is an air-based combustion technology featuring inherent separation of CO_2. It involves the use of a metal oxide as a carrier to transfer oxygen from the combustion air to the fuel. Direct contact between the fuel and the combustion air is thus avoided. As a result, the products of combustion, CO_2, and water are separated from the nitrogen and any excess oxygen. When using fuels other than natural gas, fuel-bound nitrogen and sulphur leave the reactor as NO_x and SO_2 and can be separated using conventional technologies. CLC flue-gas volume is reduced by a factor of 10 with respect to conventional air-fired combustion. **Fig. 5** is a simplified schematic of a CLC. It typically consists of two reactors in the form of interconnected fluidized beds:

1. A fuel reactor in which the metal oxide (MeO) is reduced by the fuel, producing a gas stream of primarily CO_2 and H_2O with traces of unreacted fuel according to

$$C_xH_y + (2x + 0.5y)\mathrm{MeO} \leftrightarrow x\mathrm{CO_2} + 0.5\mathrm{H_2O} + (2x + 0.5y)\mathrm{Me} \quad \dots\dots\dots\dots (1)$$

2. An air reactor in which the reduced metal oxide from the fuel reactor is oxidized with air to produce a hot metal oxide for recycling to the fuel reactor and a flue gas composed primarily of N_2 and unreacted O_2:

$$\mathrm{Me} + 0.5\mathrm{O_2} \leftrightarrow \mathrm{MeO} + \text{heat}. \quad \dots\dots\dots\dots (2)$$

The air reactor is typically a CFB, whereas the fuel reactor is a bubbling fluidized bed. A cyclone is used to separate the MeO from the excess combustion air. Pot-seals help circulate metal oxides between the two reactors. Potential leakage can be avoided by the injection of steam instead of air to fluidize and transfer the solid particles through the pot-seals (Johansson et al. 2003). The net chemical reaction over the two reactors is the same as for conventional combustion. CLC temperature is generally in the range of 800 to 1200°C (Mattisson et al. 2001). Key operating parameters for CLC include ratio of air/fuel/MeO, solid inventory, solids circulation rate between reactors, hydrodynamics of the bed, air leakage, residence time, and pressure.

Most of the CLC work has focused on burning gaseous fuels (e.g., natural gas or syngas). Solid fuels (e.g., coal or petroleum coke) are of great interest because of their low cost and abundance, but add complexity to the process. The main challenges related to solid-fuel use in CLC include increasing the reaction rate, reducing the solid residence time, improving ash separation and residue disposal, and establishing the effects of impurities on metal oxide and on the cost of gas cleanup.

One proposed method would involve gasifying solid fuels to produce syngas. The syngas could be burned in a separate CLC unit, or it could be produced in the fuel reactor directly. In the latter approach, steam and/or CO_2 would be used to gasify the fuel and fluidize the bed. Syngas would then react with the oxygen carrier. Reaction between the metal oxide and syngas is fast; fuel gasification is the rate-limiting step. The rate is likely faster than ordinary gasification because of the high concentrations of H_2O and CO_2 present in the fuel reactor (Berguerand and Lyngfelt 2008; Gao et al. 2008). In the second method, known as uncoupled CLC, copper oxide is used to release oxygen at temperatures of approximately 1000°C, allowing the solid fuel to react with oxygen directly (Mattisson et al. 2009).

Status. The main features of CLC metal oxides are active material content, selectivity toward complete oxidation, oxygen capacity, solid reactivity, regenerability, mechanical strength, lifetime, and cost. To date, more than 300 metal oxides have been tested (Johansson et al. 2006). The most promising are NiO, CuO, Mn_3O_4, and Fe_2O_3. Common support materials include Al_2O_3, TiO_2, SiO_2, ZrO, and Al_2MgO_4. Suitable metal oxide and support material combinations must be selected on the basis of the fuel composition and process temperatures. Also, some natural materials that are a source of low-cost oxygen carriers have been studied (Pröll et al. 2009).

Currently, European universities are the main contributors to the development of CLC technology. The EU GRACE Project has resulted in the development of a Ni-based metal oxide and the piloting of a 10 kWth CLC unit for gaseous fuels with continuous solid circulation. The CO_2-Capture Project, established in 2000, is a collaboration of major international energy corporations that intends to advance CCS technology. Phase 1 studied the possibility of retrofitting refinery heaters and boilers with oxyfuel combustion and concluded that for existing refineries, large centralized ASU and FGR may result in lower CO_2-capture costs. However, in new builds, or for single-unit applications (e.g., boilers for steam extraction of heavy oil or bitumen), CLC has great potential to become the more economical option. Phase 2 (2004–09) focused on the scaleup of the CLC pilot unit at the Technical University of Vienna to 120 kW. This unit is designed for gaseous fuels and uses synthetic MeO (Miracca et al. 2009).

The Korean Institute of Energy Research began developing metal oxides in 2000 and has since then studied the kinetics and hydrodynamics of CLC using fixed and fluidized bed reactors at 50 kW_{th} (Ryu et al. 2004). Alstom Power Incorporated and Ohio State University are studying the CLC retrofit of existing coal-fired power plants. Alstom is using chemical looping of calcium oxides and is building a 3-MW_{th} unit (Abdulally et al. 2010). Ohio State University has a 25-kW_{th}-CLC unit with an iron-based MeO (Fan 2010).

In Canada, the University of British Columbia has studied CLC in a cold-flow CFB and used the results to design a hot-flow unit that is under construction. Operational parameters (e.g., conversion, pressure drop, bed expansion, solid holdup, and gas concentrations) will be studied in the new setup at temperatures of approximately 900°C.[1] The University of Western Ontario is

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[1] Ellis, N. 2009. Personal communications, University of British Columbia, Vancouver, British Columbia.

TABLE 4—SUMMARY OF TECHNOECONOMIC PARAMETERS OF CO_2-CAPTURE TECHNOLOGIES (WOODS ET AL. 2007; CIFERNO 2008)

	Coal Precombustion[a,e]	Coal Post-Combustion[b,e]	Gas Post-Combustion[c,f]	Coal Oxyfuel[d,e]
Gross output (MWe)	720	666	520	792
Auxiliary power demands (MWe)	184	118	38	246
Auxiliary power increase due to capture (%)	50%	292%	288%	720%[g]
Net output (MWe)	536	548	482	546
Net heat rate, higher heating value (HHV) (MJ/kWhe)	11.2	13.2	8.2	12.7
Heat rate increase due to capture (%)	23%	45%	16%	40%[g]
Net efficiency, HHV (%)	32.1%	27.2%	43.7%	28.3%
Efficiency loss due to capture (% points)	7.4	12.3	7.1	11.2[g]
Overnight total plant cost (USD/$kW_{\text{Net output}}$)	2529	2857	1172	2930
Cost increase due to capture (%)	36%	83%	111%	87%[g]
CO_2 capture cost (USD/tonne)	30	41	70	38

[a] Average of GE, Shell, and E-Gas gasifiers, 2 GE 7FA turbines, 3 pressure reheat heat recovery steam generator (HRSG), Claus sulphur removal, 90% CO_2 capture via Selexol scrubbing

[b] Steam cycle: 24.1 MPa/599°C/621°C, includes flue gas desulphurization (FGD) and selective catalytic reduction (SCR) flue gas treatment, 90% CO_2 capture via monoethanolamine (MEA) scrubbing

[c] 2 GE 7FA turbines, 3 pressure reheat HRSG, includes SCR flue gas treatment, 90% CO_2 capture via MEA scrubbing

[d] Steam cycle: 24.1 MPa/599°C/621°C, includes FGD flue gas treatment, 95% CO_2 capture via oxyfuel combustion with 95% (mole) pure O_2

[e] Dry coal properties are: 10.91% ash, 39.4% volatile matter, 49.7% carbon, 2.5% sulphur and 30.5 MJ/kg HHV

[f] Natural gas HHV = 52.9 MJ/kg

[g] Reference plant is an air-fired coal power plant with an identical steam cycle, but without capture

currently developing MeOs with a focus on optimizing kinetics and reactivity. The simultaneous development of proprietary software to simulate CLC processes at this institution is ongoing (Hossain et al. 2007).

Future Prospects. CLC is less mature than other capture technologies; it has a longer path to commercialization. The results from MW_{th}-scale units will support further scaling up. Process modifications, selection of suitable MeO, effect of fuel impurities and CO_2 stream purity, and emissions control are the main focus of research and development of CLC. The modification of conventional CFB reactor designs to suit CLC requirements may further advance commercialization. Establishing commercial oxygen-carrier production is needed for CLC to reach commercialization.

Further testing of existing oxygen carriers is required to establish (1) their chemical integrity after undergoing several oxidation and reduction cycles and (2) the effect of fuel-bound impurities. Oxide lifetime depends on the reactivity loss caused by morphological changes. The production of fine particles reduces the lifetime by attrition or degradation and results in fines carryover by the exhaust gas. Synthetic materials are recommended for natural-gas CLC, while low-cost natural minerals are best for coal CLC with high potential of contamination and degradation. Recent research has focused on coal (Cuadrat et al. 2010; Ksepko et al. 2010). Also, biogas and liquid fuels (Hoteit et al. 2009; Hoteit et al. 2010; Sit 2009) have been evaluated.

Summary of Technoeconomic Performance

Table 4 provides a summary of key metrics for the capture technologies presented earlier. The values shown are estimations based on the status of current technology and are meant to represent typical performance parameters of carbon capture applied to power plants. In the source study (Woods et al. 2007, Ciferno 2008), each capture technology was compared with a plant with identical configuration, but without capture, except for oxyfuel. Oxyfuel is compared with an air-fired plant featuring the same steam cycle, but without capture. For brevity, only the values corresponding to the capture designs are presented here, although the impacts of CO_2-capture implementation are shown, with respect to the no-capture plants.

When incorporated into fossil-fuel plants, capture processes affect the energy-conversion efficiency because of their additional energy requirements. Oxyfuel requires substantial energy for oxygen production in a cryogenic ASU. Its oxygen demands are higher than those of a precombustion plant with equivalent power output because the latter features only partial oxidation of the fuel, as opposed to full combustion as in the former. Post-combustion capture causes ancillary power demands to almost triple with respect to noncapture plants, though this is related mostly to CO_2 compression. Percentage-wise, precombustion suffers the lowest rise in parasitic load because the CO_2 is recovered at high pressure, requiring less energy for compression and because of its relatively lower oxygen demands.

As a consequence, more fossil fuel is required per unit of energy output because of capture. For power plants, this increases the heat rate, as observed in Table 4. Gas post-combustion power plants have the lowest increase because of their superior thermal efficiency. For coal processes, however, precombustion capture has a lower heat-rate penalty than post-combustion capture, with oxyfuel being in between. The previously described trends are also true for thermal efficiency, expressed here as percentage net efficiency points lost because of carbon capture.

Capture also imposes a cost penalty, expressed here as a percentage increase in the capital cost of each technology. On a financial basis consistent across technologies, and with respect to a no-capture plant of identical characteristics, post-combustion capture approximately doubles the cost of gas-fired plants. This is caused mainly by their flue-gas characteristics; large volumes of highly diluted CO_2 streams require large equipment, dramatically increasing capital costs. In the case of natural-gas power plants, this increase is magnified by the relatively low capital expense of a no-capture plant. The opposite is true of coal plants featuring precombustion capture; no-capture plants are capital intensive, but they share numerous components with capture plants. Hence, the additional investment required to capture CO_2 is relatively modest. Also, precombustion processes feature high CO_2 partial pressures and relatively lower gas volumes than post-combustion or oxycombustion processes, thus requiring smaller equipment.

For coal-fired processes, post-combustion or oxyfuel combustion has a similar cost impact. In the former case, large gas volumes of relatively low CO_2 concentration require large capture equipment, whereas in the latter, large oxygen demands require an ASU. However, oxyfuel technology is considerably less mature than post-combustion capture; the cost estimates presented here are thus likely to drop with future improvements in boiler design and oxygen separation, as outlined in the Oxyfuel Combustion subsection.

TABLE 5—SUITABILITY OF CO_2-CAPTURE TECHNOLOGIES ACCORDING TO OIL-SANDS PROCESS

	SAGD	Mining	Upgrading
Precombustion	Inadequate	Adequate	Best
Post-combustion	Adequate	Best	Inadequate
Oxyfuel	Best	Inadequate	Inadequate
CLC	Best	Inadequate	Adequate

In terms of capture costs, precombustion capture features the lowest per-tonne-of-CO_2-capture cost, whereas post-combustion has the highest. In the latter case, gas-fired processes have significantly higher capture costs than coal-fired ones. This is explained by Eq. 3 (Woods et al. 2007):

$$\text{Capture cost} = \frac{\text{Product cost}_{\text{w/capture}} - \text{Product cost}_{\text{w/o capture}} \left[\frac{\text{USD}}{\text{MWh}}\right]}{\text{CO}_2 \text{ removed} \left[\frac{\text{t}}{\text{MWh}}\right]} \quad \cdots\cdots\cdots\cdots\cdots (3)$$

Coal-fired processes produce substantially more CO_2 per unit of fuel than natural-gas processes. Thus, more CO_2 is captured in coal post-combustion plants, resulting in a greater denominator in Eq. 3. The difference in capture costs between coal oxyfuel and post-combustion plants is small, although costs for the former technology are not well established (ZEP 2011). Also, capture costs are heavily influenced by capital costs and the assumed fuel costs, especially for natural-gas-fired processes. The costs discussion presented here is of an introductory nature; other studies (ZEP 2011; Woods et al. 2007) offer more-in-depth analysis for the interested reader.

CO_2-Capture Integration With Oil-Sands Operations

The purpose of this section is to discuss the implementation options of CO_2-capture technologies reviewed earlier in oil-sands processes, as currently executed in Alberta. To this end, the analysis is two-fold, including a qualitative ranking of each capture technology according to its suitability of implementation in each of the major oil-sands processes (bitumen mining, SAGD extraction, and upgrading). An additional feature of the study is a detailed description of the best concepts for integrating carbon capture in oil-sands operations, from a technical-suitability perspective.

Table 5 provides an at-a-glance summary of the ranking resulting from our analyses. Each capture technology was qualitatively appraised from a suitability-of-implementation perspective. Accordingly, we describe each technology/oil-sands process combination as "best," "adequate," or "inadequate." These values reflect the overall balance of advantages and disadvantages inherent to each pair. The suitability of each capture technology is strongly dependent on individual project, process-, design- and application-specific characteristics that cannot be fully reflected in this manuscript. Also, the analyses outlined here must be complemented with a comprehensive technoeconomic analysis to reveal the financial impacts of choosing one capture technology over another for a given oil-sands project. The positioning presented here is of a high-level nature and is provided as an initial point of reference for further analyses.

Bitumen Mining. The energy demands of mining oil sands and extracting bitumen are dominated by hot-water production, power generation, and diesel fuel for trucks and mining equipment, in that order. Because only the former two CO_2 sources can be feasibly captured (e.g., trucks are mobile carbon sources), our analysis focuses on carbon-capture solutions for hot-water and power-generation processes.

Post-combustion CO_2 capture is the best fit for mining operations because it would allow the recovery of the bulk of the CO_2 produced; that is, greenhouse-gas emissions from boilers and/or cogeneration plants. Post-combustion capture could be retrofitted to existing facilities with minimal modifications to the mining process, as shown in **Fig. 6.** The main disadvantage of post-combustion capture is its large energy demands for solvent regeneration. This energy, usually in the form of steam, may be provided by the existing utility plant, provided it has excess capacity. Otherwise, increasing the capacity or building a new cogeneration plant would be required. Land is another issue that needs thorough consideration, especially in retrofit applications. Post-combustion technologies have a large footprint because they process highly diluted CO_2 streams at low pressures. Greenfield sites may be less affected by land constraints, provided that a location with adequate space for the mining operations, the capture plant, and plant utilities are available.

Gasification of hydrocarbon fuels could be used to produce power and large amounts of low-grade heat, which is compatible with bitumen mining. The gasification process yields CO_2-rich streams that can be separated by precombustion capture. Implementation of precombustion is less attractive than post-combustion capture for mining operations because of the significant cost and complexity of installing a gasifier and an ASU at a mining site that requires mostly power and hot water. Producing H_2 through gasification of bitumen or bitumen residues with CO_2 capture in an integrated mining and upgrading operation would be a more-compelling situation than for mining alone. Thus, precombustion CO_2 capture would be preferable only if natural gas was substantially more expensive than other fuels (e.g., bitumen or low/negative-value asphaltenes) that might be readily available at the site.

For greenfield mining developments, an attractive alternative would be to combine a bitumen or bitumen-residue-fired gasifier with precombustion capture and a Fischer-Tropsch process to supply power, hot water, and diesel fuel for mining equipment. This is a novel concept that requires a thorough technoeconomic evaluation, and deserves to be explored, particularly for integrated

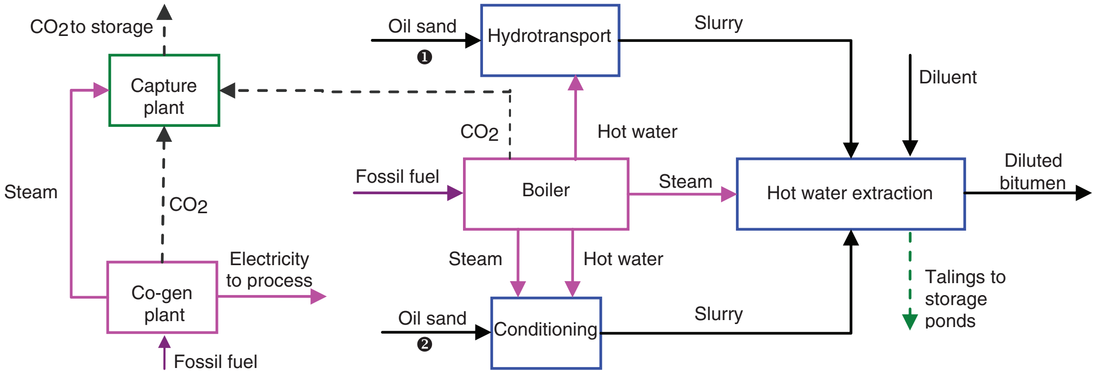

Fig. 6—Bitumen mining with post-combustion CO_2 capture.

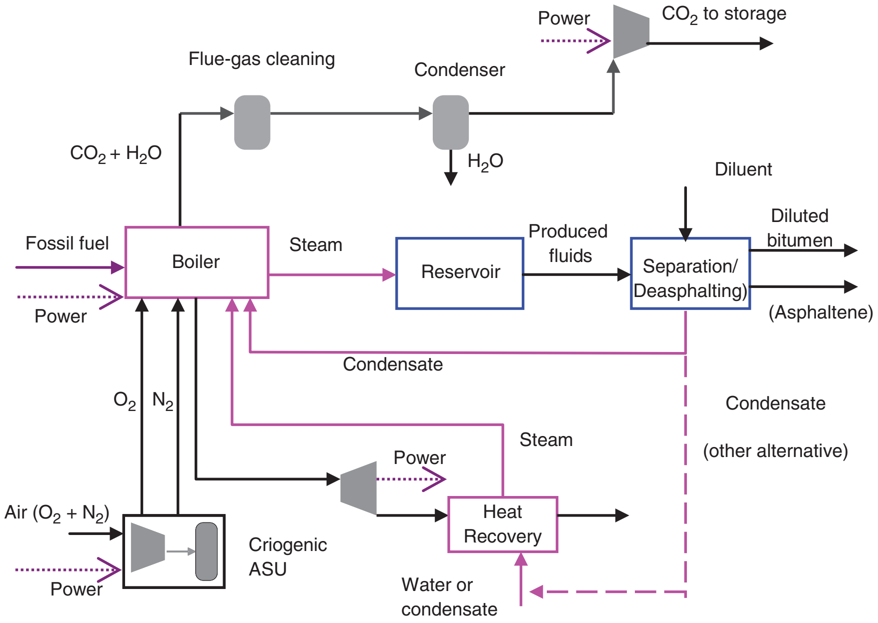

Fig. 7—SAGD with oxyfuel-combustion and ASU integration.

mining and upgrading operations, because this concept could fully supply the energy demands of the site (including H_2) while capturing most of the carbon produced.

Oxyfuel and CLC are technologies best suited for large-scale steam production and power generation and hence are suboptimal for bitumen-mining processes, where hot water is the main energy input to the process. Although it is possible to retrofit existing air-fired boilers to operate in oxyfuel mode, a new ASU would be needed, which adds a potentially unacceptable cost. Oxyfuel- or CLC-based CO_2 capture for mining operations may be more attractive if excess steam and power produced could be readily exported and sold to other users in the vicinity. Currently, most mining operations are located in remote areas with insufficient power-transmission infrastructure to connect these sites with load centres.

SAGD Bitumen Extraction. The main steps of the SAGD process are steam generation and bitumen separation. The steam is typically produced in a once-through natural-gas air-fired boiler. In the separation process, the solution gas and sometimes asphaltenes are removed from the bitumen. The energy demands of SAGD operations consist mainly of steam, with electricity accounting for less than 5% of the total energy (Ordorica-Garcia 2007).

Currently, the best option for CO_2 capture in SAGD is the oxyfuel process. Oxyfuel combustion can be feasibly applied to large-scale boilers, which are the chief source of carbon emissions in SAGD operations. Further, existing boilers could be retrofitted or replaced by new oxyboilers. Oxyfuel integration with SAGD can be accomplished in two ways: (1) by replacing the natural-gas boiler with a natural-gas combined cycle featuring an oxyfired gas turbine combustion chamber (Anderson et al. 2008) and (2) by replacing the natural-gas boiler with a fluidized bed combustor. This would allow the use of locally available opportunity fuels (e.g., bitumen or asphaltenes), if natural-gas prices are substantially higher.

One attractive option for integrating oxyfuel combustion with the steam cycle is shown in **Fig. 7,** based on the integration scheme proposed by Tranier et al. (2009). Fuel is oxyfired in a boiler, yielding a highly concentrated CO_2. Pressurized N_2 from the ASU (which would normally be vented) is heated in the boiler and expanded in a turbine, generating additional electricity to offset the efficiency loss caused by the ASU.

Adding post-combustion capture downstream of the boiler of a SAGD plant is another option. This is less attractive than oxyfuel because it would involve producing large quantities of steam for solvent regeneration, in addition to the steam required for SAGD, thus increasing CO_2 production. For retrofits, the existing boiler would be unlikely to have capacity for producing the extra steam, unless it had been oversized originally. Thus, in addition to the cost of the capture plant, a post-combustion process would incur additional costs for supplemental-steam-generation facilities.

The simplest solution would be to add a new gas boiler to generate the steam needed for solvent regeneration. This would not be a good option because producing low-pressure steam would lead to large energy losses. A cogeneration plant would be a better alternative, producing electricity in a steam turbine using high-pressure steam and supplying the required low-/medium-pressure steam for post-combustion capture. Building new facilities to supply energy for post-combustion capture depends on project-specific technoeconomic considerations (i.e., selling excess power to the grid or derating the existing utility plant). Other issues to consider are space for the capture plant and increased water consumption because of the extra steam demands.

Precombustion is not an inherently good candidate for retrofit, and would thus be limited to greenfield SAGD plants. Precombustion technology is not a good fit for SAGD operations because integrating precombustion capture with SAGD would separate the combustion and capture processes. Hence, even after CO_2 recovery downstream of the gasification process, extra CO_2 would be generated in the steam generator (because of syngas combustion at low pressure), requiring more capture equipment downstream of the boiler. The latter would largely negate the advantage of capturing CO_2 upstream of the boiler at high pressure and high concentration.

Integration of CLC with SAGD offers unique advantages over other capture technologies because of its inherent separation of CO_2 without the expense of an oxygen plant (**Fig. 8**). For this reason, in the future CLC may rival oxyfuel as the best carbon-mitigation option for SAGD operations, provided that scaling up of CLC steam boilers to industrial scales is accomplished successfully.

Bitumen Upgrading. The upgrading stage involves the conversion of bitumen from mining and/or SAGD to synthetic crude oil. As a result of its extensive energy consumption, upgrading alone is responsible for 65 to 80% of the total greenhouse-gas emissions associated with SCO production (Ordorica-Garcia 2007). Upgrading overwhelmingly requires H_2 and steam, and relatively smaller

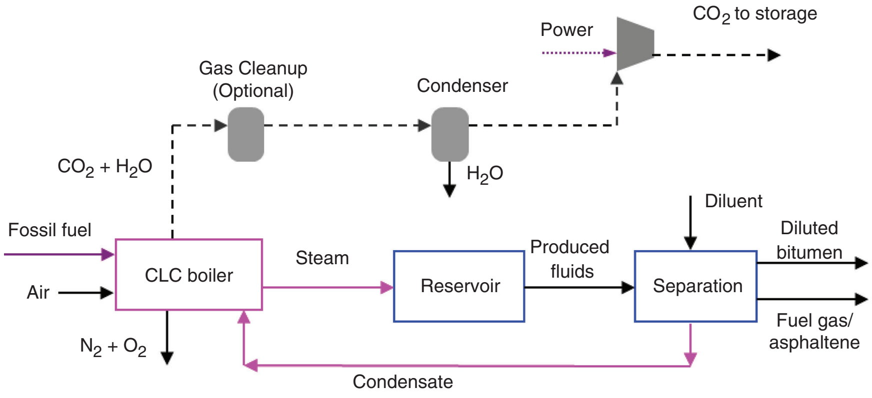

Fig. 8—SAGD with CLC.

amounts of electricity and process fuel. Hydrogen is needed for hydrotreating of light and heavy gas-oils and for hydrocracking of vacuum-topped bitumen. H_2 is typically produced through SMR, in which heat is supplied by burning some of the feed gas. The steam required for reforming is raised by cooling the reformed gas stream. After the WGS reaction, a bulk stream of CO_2 is separated using either chemical or physical absorption and vented, while the hydrogen stream is purified further using pressure-swing adsorption (PSA).

Precombustion CO_2 capture is the best alternative to reduce the carbon emissions of H_2 production in upgrading operations. Petcoke and asphaltene are byproducts of current upgrading processes that are ideal feedstock for gasification. However, they are produced in quantities insufficient to satisfy all of the H_2 demands of upgrading. Blending coal with petcoke could solve this limitation, negating the need for SMR, offsetting the use of natural gas by turning byproducts that are currently stockpiled into hydrogen. The CO_2 from precombustion capture would be available at high partial pressure, reducing compression requirements/costs and easing its transport.

The hydrogen or syngas produced through gasification could also replace fossil fuels used in other upgrading processes. Furthermore, a significant amount of useful steam and low-grade heat would be available. In principle, a gasification plant with precombustion CO_2 capture could feasibly coproduce steam, power, and hydrogen in quantities adequate to fully support upgrading operations on a large scale. This concept, known as polygeneration, is depicted in **Fig. 9.**

The application of post-combustion capture for upgrading operations is the least attractive option for two fundamental reasons: (1) the largest CO_2 source is the hydrogen plant; the CO_2 concentration downstream of the hydrogen plant and upstream of the PSA is high and therefore more suitable for capture through precombustion; (2) CO_2 sources other than the hydrogen plant (and cogeneration plant, if present) are relatively smaller and geographically dispersed. Post-combustion capture of these sources would necessitate extensive ducting of vast volumes of low-concentration flue gases from several locations to large centralized absorber units, incurring high costs and substantial energy demands for flue-gas transport. An additional difficulty with implementing post-combustion capture would be the considerable space requirements to accommodate the ducting and the absorption and regeneration columns required to process large flows of flue gases at atmospheric pressure, with fairly low CO_2 concentrations.

On its own, CO_2 capture through oxyfuel is an inadequate solution to reduce the carbon emissions of upgrading. Oxyfuel

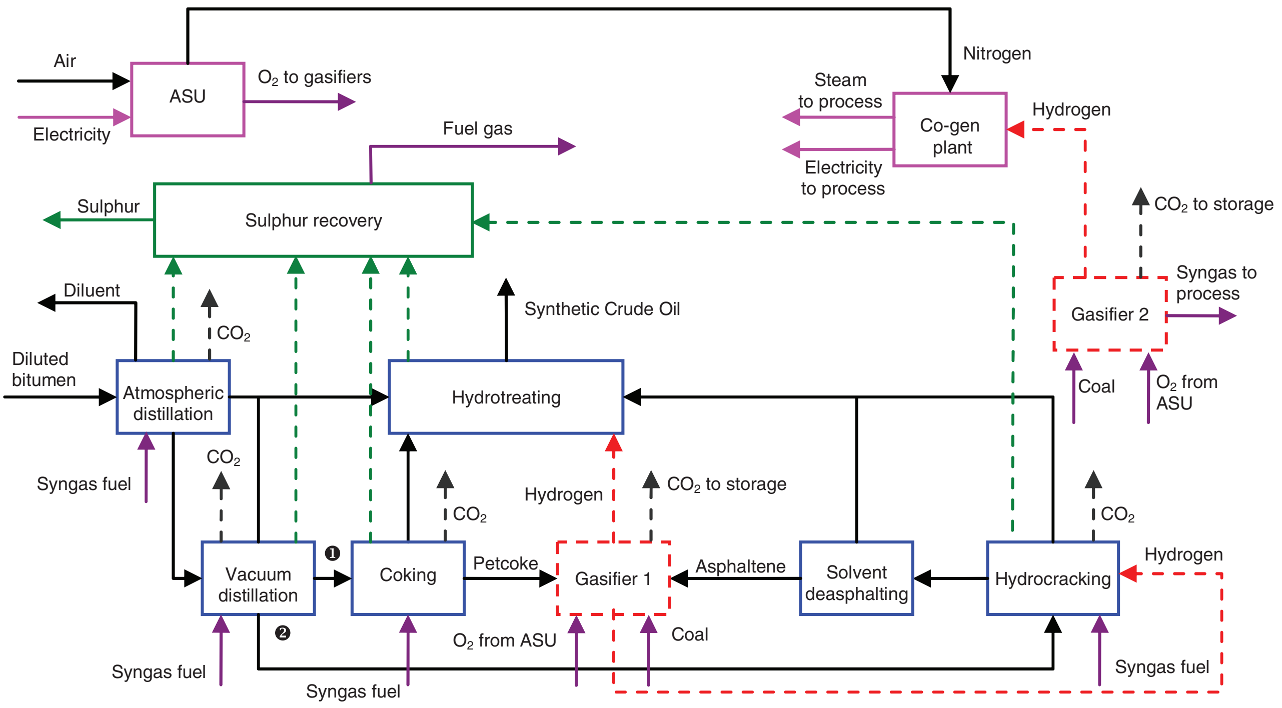

Fig. 9—Upgrading featuring polygeneration gasification with precombustion CO_2 capture.

combustion is ill-suited for incorporation with a hydrogen plant, and thus it cannot mitigate the main source of CO_2. What it may feasibly address is capturing CO_2 from the small, distributed sources in upgrading – namely, the furnaces found in the distillation columns, cokers, and crackers, in addition to cogeneration facilities. In the short term, existing natural-gas furnaces could be retrofitted for oxyfired operation, while in the longer term, bitumen-residue streams could be oxyfired in CFB combustors.

In combination with precombustion capture in the hydrogen plant, oxyfuel would enable CO_2-capture rates approximating 100% for the entire upgrading facility. A further advantage of this concept would be the potential to use a single ASU to supply all of the oxygen for the gasifier and the oxyfurnaces. A disadvantage would be the need to gather the CO_2 generated in the oxyfurnaces and oxyboilers. This would require extensive piping to gather the CO_2 for centralized compression before transport. Although the flue-gas volumes involved would be significantly smaller than in the post-combustion capture option, the CO_2-gathering system would increase the land requirements. Further study is needed to determine whether the incremental CO_2 capture attainable through oxyfuel is economically feasible.

In the future, CLC may offer a viable alternative to oxyfuel implementation for CO_2 mitigation in upgrading operations. Gas-fired CLC-based furnaces could enable the capture of CO_2 from distillation columns, crackers, and cokers. Likewise, the utilities required in upgrading could be produced in a CLC boiler. The main advantage of CLC over precombustion and oxyfuel is that no oxygen plant is needed for carbon capture, which would yield significant cost reductions.

CLC technology may also be used to gasify upgrading residues or reform natural gas with inherent CO_2 separation, effectively eliminating the need for SMR or gasification plants for hydrogen production. This concept, called chemical looping reforming (CLR) or chemical looping gasification (de Diego et al. 2009; Moghaderi and Wall 2007), may provide an alternative to precombustion capture in the long term. The main difference between CLR and CLC is that with CLR, oxygen is supplied in quantities lower than the stoichiometric combustion amount, yielding CO and H_2 instead of CO_2 and water. CLR with solid fuels is in a laboratory-scale stage of development at the time of writing.

Two things must be noted concerning the implementation of CLC in upgrading operations. First, CLC is in its early stages of development and will likely see its first implementation in SAGD applications, where steam is the main product. Second, unlike other capture technologies, CLC would not be easily retrofitable to existing upgrading facilities, meaning that it could be applied only in greenfield sites. Precombustion-based plants will likely be deployed first because some future upgraders in Alberta plan to use gasification of bitumen residues for H_2 production. Gasification inherently requires CO_2 separation to produce high-purity H_2, thus only CO_2 compression is needed to achieve real emissions reductions in bitumen upgrading.

Conclusions

This is one of the first studies addressing the integration of existing CO_2-capture technologies with current oil-sands operations. This paper is a high-level guide for future detailed assessments of carbon-capture implementation options for the oil-sands industry.

Concerning mining operations, post-combustion is the best fit to capture CO_2, both in existing and greenfield applications. For SAGD operations, oxyfuel is the best choice to achieve carbon-emissions reductions from steam generation, for retrofit and greenfield applications. Further, developments in oxyfuel technology mean that multiple approaches for oxyfuel implementation in SAGD are possible. For future SAGD projects, however, CLC may offer a superior carbon-capture choice than oxyfuel because CLC does not incur the expense of a large oxygen-separation plant.

For upgrading, precombustion capture is the best method to achieve substantial CO_2-emissions reductions by gasifying bitumen residues and coal blends to produce H_2. The entire energy needs of upgrading could be met by gasification/polygeneration with CO_2 capture. In upgrading operations, oxyfuel combustion could be used to obtain high-purity CO_2 streams from decentralized sources (e.g., furnaces and heaters), while CLR may prove to be a viable alternative for clean-hydrogen production in the future. There is no single universally superior integration option, but the combinations mentioned herein are, in the authors' perspective, the most suitable.

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