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from Integrated Gasification/Combined-Cycle Systems**

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The Potential for Control of Carbon Dioxide Emissions from Integrated Gasification/Combined-Cycle Systems

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ABSTRACT

Initiatives to limit carbon dioxide (CO₂) emissions have drawn considerable interest to integrated gasification/combined-cycle (IGCC) power generation, a process that reduces CO₂ production through efficient fuel use and is amenable to CO₂ capture. This paper presents a comparison of energy systems that encompass fuel supply, an IGCC system, CO₂ recovery using commercial technologies, CO₂ transport by pipeline, and land-based sequestering in geological reservoirs. The intent is to evaluate the energy-efficiency impacts of controlling CO₂ in such systems and to provide the CO₂ budget, or an "equivalent CO₂" budget, associated with each of the individual energy-cycle steps. The value used for the "equivalent CO₂" budget is 1 kg/kWh CO₂. The base case for the comparison is a 457-MW IGCC system that uses an air-blown Kellogg-Rust-Westinghouse (KRW) agglomerating fluidized-bed gasifier, Illinois #6 bituminous coal, and in-bed sulfur removal. Mining, preparation, and transportation of the coal and limestone result in a net system electric power production of 454 MW with a 0.835 kg/kWh CO₂ release rate. For comparison, the gasifier output is taken through a water-gas shift to convert CO to CO₂ and then processed in a glycol-based absorber unit to recover CO₂ prior to the combustion turbine. A 500-km pipeline then transports the CO₂ for geological sequestering. The net electric power production for the system with CO₂ recovery is 381 MW with a 0.156 kg/kWh CO₂ release rate.

BACKGROUND

The possibility of global climate change resulting from increasing levels of "greenhouse" gases is the subject of considerable debate and uncertainty. Because of these concerns, policies to limit CO₂ emissions are being discussed in the United States and in various international forums.¹ The options under consideration include strong energy-conservation measures, the capture and sequestering of CO₂, and the substitution of nonfossil energy sources for fossil-fuel combustion. Discussion of these issues has drawn considerable interest to power-generating systems that minimize the production of CO₂ and are particularly amenable to CO₂ capture.

One approach, which has become an increasingly attractive option among emerging technologies, combines coal gasification, gas purification, and power generation using a gas-turbine topping cycle and a steam-turbine bottoming cycle. These IGCC systems provide high energy-conversion efficiency, with the prospect of even higher efficiencies if higher-temperature turbines and hot-gas cleanup systems are developed. Second, very low emission levels for sulfur and nitrogen species have been demonstrated at such facilities as the Cool Water IGCC plant in California. Third, IGCC plants produce gas streams with concentrated CO₂ and high levels of carbon monoxide (CO), which can be easily converted to CO₂. Capture of this CO₂ prior to combustion requires the treatment of substantially smaller gas volumes than

capture after combustion. Therefore, recovery of CO₂ in IGCC systems is potentially less expensive than in conventional combustion systems. In addition, CO₂ recovery may be accomplished in conjunction with hydrogen sulfide (H₂S) removal by using several commercial technologies. Advanced process concepts now under development offer the prospect of further, significant improvements. However, the capture and disposal of CO₂ is still expected to have significant efficiency and economic impacts upon the IGCC energy system.

This paper presents interim results from a study of the impacts associated with CO₂ recovery in IGCC systems, which is being conducted for the Morgantown Energy Technology Center by Argonne National Laboratory. The objective of the study is to compare, on a consistent systems-oriented basis, the impacts of adding CO₂ capture to an IGCC system, followed by transport of the CO₂ to a geological repository for disposal. The Argonne project also includes experimental work to investigate novel techniques for utilizing CO₂, but that effort is outside the scope of this paper. The research reported here has emphasized commercial technologies for capturing CO₂, but ongoing work is aimed at evaluating advanced technologies under development and novel power-system configurations that may enhance system efficiency.

ENERGY SYSTEM

The energy system definition for the study extends from the coal mine to the final geological repository for the CO₂, as shown in Figure 1. The location of the IGCC plant is specified only as the midwestern United States. Details of the IGCC portion of the system are taken from Reference 2, which describes a plant utilizing an air-blown KRW gasifier with in-bed sulfur removal. In each case studied, the CO₂ recovery technologies have been integrated into the plant design as much as possible to limit efficiency losses. For each part of the energy system, CO₂ emissions have been either computed directly from process stream compositions or calculated from the energy consumption using a "CO₂ equivalence" of 1 kg/kWh CO₂. In this way, a total CO₂ budget for the system can be derived and compared with those for other options, thereby taking into account effects outside the immediate plant boundary.

Mining, Preparation, and Transportation of Raw Materials

Illinois #6 coal from the Old Ben No. 26 mine is the fuel used in all of the cases studied. The ultimate analysis appears in Table 1.² Old Ben No. 26 is an underground mine with an associated coal-preparation plant. The power plant is assumed to be 160 km from the mine, shipment of the coal being by rail using a unit train. In order to supply the 152,667 kg/h of coal required by the IGCC plant, the mine and preparation plant combination requires 2.42 MW of power and produces 2,418 kg/h of CO₂. Another 637 kg/h of CO₂ is produced by the diesel-powered rail transportation.

Limestone is used for in-bed sulfur capture in the gasifier. It is assumed that the limestone is extracted from a surface mine (quarry) about 160 km from the plant and transported by rail to the plant site. A total of 39,795 kg/h of limestone is used by the process, giving an energy consumption for extraction of 0.257 MW with an associated CO₂ production of 257 kg/h. Shipment of the limestone by diesel-powered train produces another 140 kg/h of CO₂.

Coal and Limestone Handling

The coal-handling system at the plant includes equipment for unloading the coal from the unit train, passing it through magnetic separators, and then conveying it to 14-h storage silos. The coal is crushed and dried in a series of three fluidized-bed roller mills. The heat for drying is provided by the hot

(760 °C) flue gas from the IGCC sulfator process. This drying results in a significant CO₂ emission from the energy cycle that is not reclaimed, but presents a possible opportunity for further reductions. The coal is then held in a 2-h bunker, from which it is pneumatically conveyed to surge bins ahead of the gasifier lock hoppers. The CO₂ emissions from the sulfator are 13,099 kg/h. Limestone is also prepared by crushing it in two pulverizers and is then pneumatically conveyed to a 24-h storage silo and a 2-h storage bunker before being mixed with the coal in the gasifier surge bins. Energy consumption in this part of the plant is 3.58 MW.

Integrated Gasification Combined-Cycle Base Case

The base case for the comparisons uses air-blown KRW fluidized-bed gasifiers and in-bed sulfur removal.² A simplified schematic for this process appears in Figure 2. The system includes two heavy-duty industrial gas turbines (1260 °C firing temperature) coupled with a reheat steam-turbine bottoming cycle. Spent limestone and ash from the gasifier are oxidized in an external sulfator prior to disposal. As noted previously, the sulfator flue gas is taken to the coal-preparation operation for drying coal and is not integrated into the later CO₂ recovery operation, nor is the small amount (158 kg/h) of CO₂ that is emitted from the gasifier lock hoppers.

The hot-gas cleanup system for particulate matter consists of a cyclone followed by a ceramic-candle-type filter. Solids collected are sent to the external sulfator prior to disposal. Gas temperatures are maintained at approximately 540 °C. Supplemental hot-gas desulfurization is accomplished in a fixed-bed zinc-ferrite system. Off-gas from the regeneration of this polishing step is recycled to the gasifier for in-bed sulfur capture.

The balance of the in-plant consumption of electricity is 33.0 MW, with a main stack CO₂ emission of 362,820 kg/h. The gross IGCC electric power output is 493.8 MW; the total in-plant consumption is subtracted to yield 457.2 MW at the busbar.

Integrated Gasification Combined-Cycle with CO₂ Recovery

Several changes were made to the base case IGCC plant in order to incorporate CO₂ recovery, as shown in Figure 3. These changes entailed processing the cleaned fuel gas through a "shift" reaction to convert the CO to CO₂, recovering the CO₂, and then combusting the low-CO₂ fuel gas in a modified turbine/steam cycle to produce electricity. Gas cleaning and sulfator performance were considered to be unaffected by these changes.

The fuel gas from the KRW process is high in CO. Conversion of the CO to CO₂ in the combustion process would result in considerable dilution of the resulting CO₂ with nitrogen from the combustion air and with water from the combustion reaction. If the CO₂ is removed prior to combustion, a substantial savings in the cost of the CO₂ recovery system is possible because of reduced vessel size and solvent flow rate. The CO in the fuel gas must first be converted to CO₂ by the shift reaction:

The resulting CO₂ can then be recovered, leaving a hydrogen-rich fuel for use in the gas turbine.

The shift reaction is commonly accomplished in a catalyst-packed tubular reactor. A relatively low-cost iron-oxide catalyst is effective in the temperature range of 340 to 590 °C. Below that range, a more expensive copper-oxide catalyst is required. While the equilibrium concentrations of products are

favorable at lower temperatures, this consideration must be balanced against the need for larger reactors. In view of these conflicting considerations, high CO₂ recovery is best achieved by staged reactors that allow for cooling between stages. A two-stage system was chosen for this study, configured to achieve 95% conversion of CO to CO₂.

A number of CO₂-removal technologies are commercially available for application to IGCC systems. However, all of these options involve cooling or refrigerating the gas stream, with an attendant loss of thermal efficiency. To minimize the loss, the heat removed during cooling must be recovered and integrated into the system. Several options for this integration were evaluated, including steam generation alone, fuel-gas preheating with supplemental steam generation, and fuel-gas saturation and preheating. In the last case, which was the one chosen for this study, moisture condensed from the fuel gas prior to CO₂ recovery is injected into the clean fuel-gas stream as it is heated by recovered heat following CO₂ removal. This allows additional heat to be absorbed before combustion and increases the mass flow rate through the gas turbine. The balance of the thermal energy is used in the heat-recovery steam generator for feedwater heating and steam generation.

The commercial CO₂ recovery processes operate by absorption of the CO₂ in a liquid solvent and subsequent regeneration of the solvent to release the CO₂. The temperature of absorption is solvent-specific. In general, however, the solvents have low boiling points so that substantial cooling of the synthesis gas is required, as noted above. Furthermore, lower temperatures favor absorption, thereby reducing the necessary solvent flow rate. This implies a need for further cooling or refrigeration of the solvent, with additional energy losses. The regeneration of the solvent is also energy-intensive for most processes, since it is usually accomplished by flashing (pressure reduction) and/or heating. If flashing is employed, repressurization of the solvent is required. Heating is generally accomplished by the extraction of steam from the steam cycle.

Five alternative CO₂-recovery processes have been evaluated to date: (1) high-pressure (1000-psi) absorption by a glycol-type solvent with regeneration by flashing, (2) low-pressure (250-psi) absorption by a glycol-type solvent with regeneration by flashing, (3) absorption by monoethanolamine (MEA) with regeneration by throttling and heating via a reboiler, (4) absorption by hot potassium carbonate with regeneration by throttling and heating, primarily by a reboiler, and (5) absorption by low-temperature methanol with regeneration by thermal stripping and distillation. From an energy perspective, the glycol options have the advantage. This is particularly true for the low-pressure system, which does not require compression of the synthesis gas prior to absorption. The results presented here are those for the low-pressure glycol system.

Pipeline Transport of CO₂

Once the CO₂ has been recovered from the fuel-gas stream, its transportation, utilization, and/or disposal remain as significant issues. Carbon dioxide represents a large-volume, relatively low-value by-product that cannot be sequestered in the same way as most coal-utilization wastes (i.e., by landfilling). Large volumes of recovered CO₂ are likely to be moved by pipeline. In some cases, existing pipelines could be used, perhaps in a shared mode with other products. In other cases, new pipeline construction would be required. Costs for pipeline construction and use vary greatly on a regional basis within the United States.

The recovered CO₂ (328,810 kg/h) represents about four million normal cubic meters per day (4×10^6 Nm³/d) of gas volume. A survey of 13 natural gas pipeline projects³ was used to determine the

relationship between installed compressor capacity, gas volume moved, and pipeline length. The initial compression of the gas to pipeline pressures constitutes the major source of power consumption, amounting to about 33 MW of additional use within the IGCC plant. The receptor site for sequestering CO₂ is assumed to be 500 km from the plant site, and pipeline booster stations are expected to consume another 2.07 MW with an equivalent CO₂ release of 2066 kg/h.

CO₂ Sequestering

It has been proposed that CO₂ be disposed of in the ocean depths. However, many engineering and ecological concerns associated with such an option remain unanswered, and the earliest likely reservoir is a land-based geological repository.⁴ A portion of the CO₂ can be utilized for enhanced oil recovery (EOR), which sequesters a portion of the CO₂, or it can be completely sequestered in depleted gas/oil reservoirs and nonpotable aquifers. Both the availability of these zones and the technical and economic limits to their use need to be better characterized. The energy required at the sequestering site was analyzed, recognizing that the power required for compression will rise throughout the life-cycle of these sequestering reservoirs. However, it was found that the first reservoirs that would be used will in fact be capable of accepting all IGCC CO₂ gas for a 30-yr period without requiring any additional compression costs for operation.

ENERGY CYCLE

Comparisons of the system CO₂ and energy balances are given in Tables 2 and 3, respectively. The amounts of raw materials (coal and limestone) supplied to the plant are assumed to be constant for both the base and CO₂-recovery cases. However, gross power generation varies with changes in internal energy use (primarily affecting the steam turbine output). Energy use for plant auxiliaries and the CO₂ recovery process is subtracted from the gross generation to yield the net busbar output. This must be further reduced by the external power consumption to yield the net system energy output.

For the recovery case, the system efficiency is reduced from the base-case value of 39.5% to 33.5% with a reduction of about 74 MW in the net system output. If the energy used for compression of the CO₂ at the IGCC plant is combined with the booster-station requirements, nearly one-half of that reduction is seen to be due to transportation of the CO₂ to the disposal site. Nevertheless, the total CO₂ emissions are reduced to only 16% of those in the base case, and the emission rate is reduced from 0.835 kg/kWh to only 0.156 kg/kWh. It should be noted that replacement of the "lost" 74 MW would likely entail additional CO₂ emissions that are not accounted for here. The incremental capital and operating costs associated with CO₂ recovery have not been fully evaluated as yet, but the loss in efficiency alone will produce a significant increase in electricity costs.

ACKNOWLEDGMENT

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REFERENCES

1. Princiotta, F.T., 1992, "Greenhouse Warming: The Mitigation Challenge," Proceedings of the 1992 Greenhouse Gas Emissions and Mitigation Research Symposium, sponsored by the U.S. Environmental Protection Agency, Washington, D.C., Aug. 18-20.

2. Southern Company Services, Inc., et al., 1990, *Assessment of Coal Gasification/Hot Gas Cleanup Based Advanced Gas Turbine Systems*, DOE/MC/26019.3004 (DE91002084), prepared by Southern Company Services, Inc., Birmingham, Ala., for the U.S. Department of Energy, Morgantown Energy Technology Center, Morgantown, W.Va.
3. Guldman, J.M., 1990, "Natural Gas Market Expansion and Delivery Infrastructure Costs: Case Study for New England," ANL/EAIS/TM-21, p. 231, Argonne National Laboratory, Argonne, Ill.
4. Hangebrauck, R.P., R.H. Borgwardt, and C.D. Geron, 1992, "Carbon Dioxide Sequestration," Proceedings of the 1992 Greenhouse Gas Emissions and Mitigation Research Symposium, sponsored by the U.S. Environmental Protection Agency, Washington, D.C., Aug. 18-20.

Table 1. Coal Analysis: Illinois #6 Seam, Old Ben No. 26 Mine.²

Component	Ultimate Analysis	
	As-received (wt%)	Properties
Moisture	11.12	Ash Fusion (reducing conditions) (°C)
Carbon	63.75	Initial deformation 1,201
Hydrogen	4.50	Softening (H=W) 1,238
Nitrogen	1.25	Softening (H=1/2W) 1,285
Chlorine	0.29	Fluid 1,324
Sulfur	2.51	
Ash	9.70	Higher heating value (J/kg) 27.13 x 10 ⁶
Oxygen (by diff.)	6.88	
TOTAL	100.0	

Table 2. Comparison of CO₂ Emissions.

System Component	Base-Case CO ₂ Emissions (kg/h)	Recovery-Case CO ₂ Emissions (kg/h)
Coal supply	3,055	3,055
Limestone supply	397	397
<u>IGCC system</u>		
Coal/limestone prep.	13,099	13,099
Gasifier island	158	158
Power island	362,820	362,820
CO ₂ recovery	-	(328,810)
Subtotal	376,077	47,267
<u>Pipeline disposal</u>		
Pipeline input	-	328,810
Booster stations	-	2,066
Reservoir (2% loss)	-	(322,234)
Subtotal	-	8,642
SYSTEM TOTAL	379,529	59,361

Table 3. System Energy Balance Comparison.

System Component	Energy (MW)	
	Base Case	CO ₂ Removal
Coal supply use	(2.52)	(2.52)
Limestone supply use	(0.28)	(0.28)
<u>IGCC system use</u>		
Coal/limestone prep.	(3.58)	(3.58)
Gasifier island	(21.66)	(21.66)
Power island	(11.39)	(11.39)
CO ₂ recovery	-	(17.66)
CO ₂ compression	-	(33.06)
Subtotal	(36.63)	(87.35)
<u>IGCC system generation</u>		
Gas turbine	311.60	281.60
Steam turbine	182.20	191.40
Total	493.80	473.00
NET PLANT OUTPUT	457.17	385.65
Pipeline/disposal use	-	(2.07)
NET SYSTEM OUTPUT	454.37	380.78

Figure 1. Energy system components for CO₂ recovery.

Figure 2. Block diagram of the base case IGCC system.

Figure 3. Block diagram of the IGCC system with CO₂ recovery

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