CONF. 9: ANL/ES/CP--78143

DE93 009971

# GASIFICATION COMBINED CYCLE: CARBON DIOXIDE RECOVERY, TRANSPORT, AND DISPOSAL<sup>1</sup>

R.D. Doctor, J.C. Molburg, P. Thimmapuram, G.F. Berry, C.D. Livengood, and R.A. Johnson<sup>2</sup>

Argonne National Laboratory, 9700 South Cass Avenue Argonne, Illinois, 60439, U.S.A.

## ABSTRACT

Initiatives to limit carbon dioxide (CO<sub>2</sub>) emissions have drawn considerable interest to integrated gasification combined-cycle (IGCC) power generation. This process can reduce CO<sub>2</sub> production because of its higher efficiency, and it is amenable to  $CO_2$  capture, because  $CO_2$  can be removed before combustion and the associated dilution with atmospheric nitrogen. This paper presents a process-design baseline that encompasses the IGCC system, CO<sub>2</sub> transport by pipeline, and land-based sequestering of  $CO_2$  in geological reservoirs.

The intent of this study is to provide the CO<sub>2</sub> budget, or an "equivalent CO<sub>2</sub>" budget, associated with each of the individual energy-cycle steps. Design capital and operating costs for the process are included in the full study but are not reported in the present paper. The value used for the "equivalent  $CO_2$ " budget will be 1 kg  $CO_2/kWh_e$ .

The base case is a 470-MW (at the busbar) IGCC system using an air-blown Kellogg Rust Westinghouse (KRW) agglomerating fluidized bed gasifier, U.S. Illinois #6 bituminous coal feed, and in-bed sulfur removal. Mining, feed preparation, and conversion result in a net electric power production of 461 MW, with a 0.830 kg/kWh<sub>e</sub> CO<sub>2</sub> release rate. In the CO<sub>2</sub> recovery case, the gasifier output is taken through water-gas shift and then to Selexol, a glycol-based absorber-stripper process that recovers CO<sub>2</sub> before it enters the combustion turbine. This process results in 350 MW at the busbar. A 500-km pipeline takes the recovered  $CO_2$  to geological sequestering. The net electric power production in the recovery case is 320 MW, with a 0.234 kg/kWh<sub>e</sub> CO<sub>2</sub> release rate.

## **KEYWORDS**

CO<sub>2</sub> emissions; power production; coal gasification; integrated gasification combined-cycle; CO<sub>2</sub> removal; CO<sub>2</sub> transport; CO<sub>2</sub> sequestering.

de MASTERIA COMPANY

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

Work supported by the U.S. Department of Energy, Assistant Secretary for Fossil Energy, under contract W-31-109-Eng-38.

Affiliated with the Morgantown Energy Technology Center, 3610 Collins Ferry Rd., Morgantown, West 2 Virginia 26507, U.S.A.

# BACKGROUND

The possibility of global climate change resulting from increasing levels of "greenhouse" gases is the subject of considerable debate and uncertainty. However, the increasing atmospheric concentration of carbon dioxide ( $CO_2$ ) could have significant impacts that may not be easily reversed. Because of these concerns, policies to limit  $CO_2$  emissions are being discussed both in the United States and in various international forums (Princiotta, 1992). The options under consideration include calls for strong energy-conservation measures, the capture and sequestering of  $CO_2$ , 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  $CO_2$  and are amenable to  $CO_2$  capture.

Integrated gasification combined-cycle (IGCC) systems, which are becoming an increasingly attractive option among the emerging technologies, combine several desired attributes. First, 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 flue-gas streams with concentrated  $CO_2$ , as well as high levels of carbon monoxide (CO), which can be easily converted to  $CO_2$ . Capture of this  $CO_2$  prior to combustion requires the treatment of substantially smaller gas volumes than capture after combustion (the method that would be required with direct coal-firing), because the fuel-gas stream is not yet diluted with atmospheric nitrogen(N<sub>2</sub>) and excess air.

Recovery of  $CO_2$  in IGCC systems is potentially less expensive than in conventional combustion systems.  $CO_2$  recovery now can be done in conjunction with hydrogen sulfide (H<sub>2</sub>S) removal by using several commercial technologies. At the same time, advanced process concepts now under development offer the prospect for further significant improvements.

This paper presents a process-design baseline that encompasses the IGCC system,  $CO_2$  recovery,  $CO_2$  transport by pipeline, and land-based sequestering of  $CO_2$  in geological reservoirs. The intent of this study is to provide the  $CO_2$  budget, or an "equivalent  $CO_2$ " budget, associated with each of the individual energy-cycle steps. The base case chosen is a 470-MW IGCC system using an air-blown Kellogg Rust Westinghouse (KRW) gasifier and in-bed sulfur removal.

#### MINING

Illinois #6 coal provides the fuel for the base-case study. Its ultimate analysis appears in Table 1. The location of the IGCC plant is specified only as the midwestern United States. To investigate the significance of coal mining and transportation on the fuel-cycle  $CO_2$  emissions, emission rates typical of the U.S. were used (Marlund, 1983). These emission rates appear in Table 2, and the low extraction energy penalties, compared to other fossil fuels, reflect both the abundance of coal and the ease of recovering it. The IGCC plant uses 128,507 kg/h of the design coal, resulting in an emission of 6,689 kg/h  $CO_2$  from the mining operation and 100-km rail transport. The power consumption is 5.66 MW. A total of 14,816 kg/h of limestone is used by the process. Assuming the same extraction energy use as for coal, the use of limestone yields 772 kg/h  $CO_2$  and requires 0.59 MW of power. The total electric power requirement is 6.25 MW.

Ultimate analysis	As-received (wt%)	Ash fusion temperature	Reducing (°C)
Moisture	11.12	Initial deformation	1,201
Carbon	63.75	Softening $(H = W)$	1,238
Hydrogen	4.50	Softening $(H = 1/2W)$	1,285
Nitrogen	1.25	Fluid	1,324
Chlorine	0.29		
Sulfur	2.51		
Ash	9.70	Higher heating value (J/kg)	$27.11 \times 10^{\circ}$
Oxygen (by difference)	6.88		
TOTAL	100.0		

 Table 1. Base coal analysis:
 Illinois #6 seam, Old Ben no. 26 mine (data from Southern Company Services, 1990).

Table 2. Typical  $CO_2$  emissions for U.S. fossil fuel energy use (data from Marlund, 1983).

Fuel	Extraction, Production, and Transportation (kg CO <sub>2</sub> /J×10 <sup>9</sup> )	End-use (kg CO <sub>2</sub> /J×10 <sup>9</sup> )	Total (kg CO <sub>2</sub> /J×10 <sup>9</sup> )	
Natural gas	9.37	49.9	59.3	
Oil	8.41	72.2	80.6	
Coal	1.92	89.1	91.0	
Synthetic oil	53.1	72.2	125.2	
Synthetic gas	103.0	49.9	152.9	
Shale Oil	101.1	72.2	173.2	

### COAL/LIMESTONE PREPARATION

The coal and limestone handling system assumes unloading from a unit train to vibrating feeders that pass the material through magnetic separation before belt conveyors unload it into a series of 14-h storage silos. The coal is crushed and dried in a series of three fluidized-bed roller mills. (A fourth unit is available as a spare). Drying is accomplished by the hot flue gas (760°C) from the IGCC sulfator process. Drying results in a significant  $CO_2$  emission that is not reclaimed in this study. However, this emission presents a good 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. Limestone is prepared by crushing it in two pulverizers. Like the coal, it is pneumatically conveyed to a 24-h storage silo and a 2-h storage bunker, and it is mixed with the coal in the gasifier surge bins. The power requirement for this system, 18.3 W-h/kg coal, is consistent with available design reports (Miller, 1985; Condorelli *et al.*, 1991). The power consumption for the preparation operation emits 14,857 kg/h  $CO_2$ , while consuming 2.6 MW.

# INTEGRATED GASIFICATION COMBINED CYCLE BASE CASE

The base case chosen is a 458-MW IGCC system using air-blown KRW fluidized-bed gasifiers and inbed sulfur removal. Detailed consideration of the design and costs for IGCC systems appears in a recent U.S. Department of Energy, Morgantown Energy Technology Center report (Southern Company

Services *et al.*, 1990). A simplified schematic for this process appears in Fig. 1. The system includes two heavy-duty industrial gas turbines  $(1,260^{\circ}C \text{ firing temperature})$ , coupled with one reheat steam turbine bottoming cycle. Spent limestone and ash from the gasifier are oxidized in an external sulfator prior to disposal. The sulfator flue gas is taken to the coal preparation operation for drying coal and is not integrated into the later CO<sub>2</sub> recovery operation.

The hot-gas cleanup system for particulates consists of a cyclone followed by a ceramic-candle-type filter. Solids collected are sent to the external sulfator before disposal. Gas temperatures are maintained at approximately 540°C. Supplemental hot-gas desulfurization is accomplished by 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 total in-plant consumption of electricity is 35.4 MW, with a main stack CO<sub>2</sub> emission of 362,565 kg/h. The gross IGCC electric power output is 505.0 MW, and the in-plant consumption is subtracted to yield 469.6 MW at the busbar.

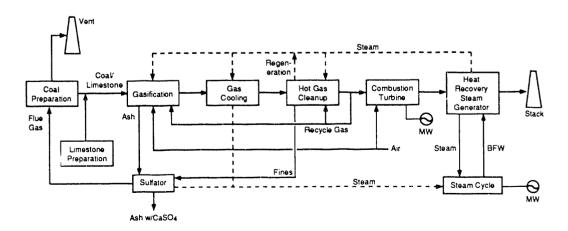
# INTEGRATED GASIFICATION COMBINED CYCLE WITH SELEXOL CO<sub>2</sub> RECOVERY

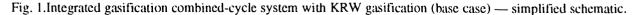
The base case described above was modified so that Selexol  $CO_2$  recovery (a process licensed by Union Carbide) could be incorporated into it. This entailed taking the cleaned fuel gas produced in gasification through a "shift" reaction to convert the CO to  $CO_2$ , recovering the  $CO_2$  by Selexol, and then combusting the low  $CO_2$  fuel gas in a modified turbine/steam cycle to produce electricity. Gas cleaning and sulfator performance were considered to be unaffected by these changes. A simplified schematic for this process appears in Fig. 2.

#### Shift Reactor

The synthesis, or "fuel" gas from the KRW process is high in CO. Conversion of the CO to  $CO_2$  in the combustion process would result in the substantial dilution of the resulting  $CO_2$  with nitrogen from the combusting air, as well as with excess air. If carbon is removed from the fuel gas prior to combustion, a substantial savings in the  $CO_2$  recovery system cost is possible because of reduced vessel size and solvent flow rate. The CO in the fuel gas must first be converted to  $CO_2$  by the shift reaction:

$$CO + H_2O ==> CO_2 + H_2$$





is internetly and the second second

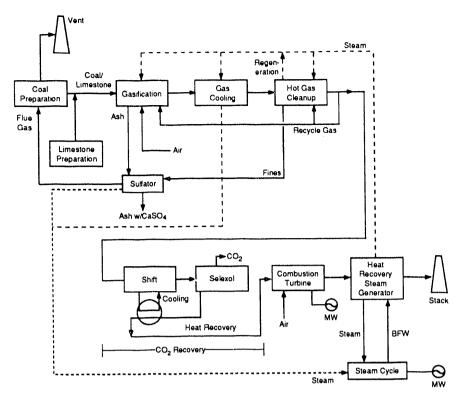


Fig. 2. Integrated gasification combined-cycle system with Selexol CO<sub>2</sub> recovery — simplified schematic.

The resulting  $CO_2$  is then recovered by the Selexol process, 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  $340^{\circ}$ C -  $590^{\circ}$ C. Below that temperature, a copper oxide catalyst is required, at much higher unit cost. While there is a favorable equilibrium concentration of products at a lower temperature, this consideration must be balanced against the need for larger reactors at lower temperature. In view of these conflicting considerations, high CO<sub>2</sub> recovery is best achieved by staged reactors that allow for cooling between stages. A two-stage system was chosen for this study, and cooling of the high-temperature shifted gas employs gas-to-gas exchange against the low-CO<sub>2</sub> fuel-gas stream from the Selexol system.

#### Carbon Dioxide Recovery by Selexol

A number of  $CO_2$ -removal technologies are commercially available for application to IGCC systems, including Selexol. However, all of these options involve cooling or refrigerating the gas stream, with an attendant loss of thermal efficiency. The Selexol glycol-based recovery process has been used at the Cool Water IGCC plant to separate H<sub>2</sub>S from CO<sub>2</sub>. Both sulfur and saleable CO<sub>2</sub> can be produced. The Selexol process uses a glycol-based solvent to strip out both CO<sub>2</sub> and H<sub>2</sub>S and then recovers them as two separate streams.

In this IGCC application, heat exchangers are used to cool the shifted fuel gas, which then is diverted and fed upward through an absorber column, where it contacts a counter-current flow of chilled lean Selexol solvent. (This is a glycol-based solvent not unlike automobile radiator fluid.) Rich solvent from the absorber is regenerated by reducing pressure to recover a  $CO_2$  product. Regenerated (or lean) Selexol solvent is then successively cooled by heat exchange with rich solvent and by supplemental ammonia refrigeration so that it can be recycled to the absorber. The product  $CO_2$  from this system is of suitable quality for long-distance pipeline transport (typically less than 2 ppm  $H_2S$ ). The flow rate of the CO<sub>2</sub> product stream to the transport pipeline is 328,995 kg/h.

#### Turbine/Steam Cycle and Plant Performance

With the shift and Selexol processes added to the IGCC system, the total in-plant consumption of electricity comes to 53.68 MW, with a main stack  $CO_2$  emission of 33,923 kg/h — a 90.9% reduction from the base case. The gross IGCC electric power output now is 403.4 MW, and the in-plant consumption is subtracted to yield 349.7 MW at the busbar.

# CO<sub>2</sub> PIPELINE TRANSPORT

Once the  $CO_2$  has been recovered from the fuel-gas stream, its transportation, utilization, and/or disposal remain as significant issues. It 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  $CO_2$  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  $CO_2$  (328,995 kg/h) represents 4.02 million normal cubic meters per day of gas volume. A survey of 13 natural gas pipeline projects (Guldman, 1990) illustrates the relationship among installed compressor capacity, gas volume moved, and pipeline length (see Fig. 3). The initial compression of the natural gas to pipeline pressures constitutes the major power consumption that must be borne by every pipeline. The receptor site for sequestering  $CO_2$  from the IGCC plant is assumed to be 500 km from the Illinois coal basin site, and a reasonable value for the energy costs to transport this gas is 10 kW/km/Nm<sup>3</sup>×10<sup>6</sup>/day. This value represents an energy consumption of 21.24 MW and an additional emission of 21,240 kg/h of  $CO_2$  (if  $CO_2$  sequestering is taking place).

# CO<sub>2</sub> SEQUESTERING

Proposals have been made to dispose of the  $CO_2$  in the ocean depths. However, many engineering and ecological concerns associated with such options remain unanswered, and the earliest likely reservoir

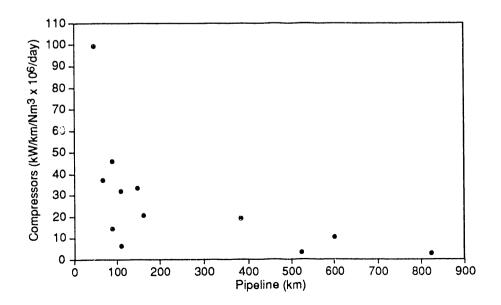


Fig. 3. CO<sub>2</sub> pipeline energy costs (data from Guldman, 1990).

is a land-based geological repository (Hangebrauck *et al.*, 1992). A portion of the  $CO_2$  can be utilized for enhanced oil recovery (EOR), 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. Levelized costs were prepared recognizing that the power required for compression will rise throughout the life cycle of these sequestering reservoirs. The first reservoirs to be used would in fact be capable of accepting all  $CO_2$  gas from IGCC systems for a 30-year period without requiring any additional compression costs for operation.

## CONCLUSIONS: ENERGY CYCLE COMPARISONS

The base case energy cycle balance for an IGCC system appears in Table 3; the IGCC with Selexol appears in Table 4. Although the addition of the Selexol process reduces  $CO_2$  emission by 90.9%, the net electric power production is also reduced by 149.5 MW. At the same time, the net greenhouse  $CO_2$  emission rate for an IGCC system with the Selexol process is 29% of the base case.

	Power (MW)	CO <sub>2</sub> Emissions (kg/h)	
Coal mining	5.66	6,695	
Limestone mining	0.59	772	
Coal/limestone rep./sulfator	2.60	12,238	
IGCC	35.4	362,822	
TOTAL	44.25	382,527	
Gross IGCC electric power	505.02		
Net IGCC power at busbar	469.62		
Net electric power with energy cycle costs	460.77		
CO <sub>2</sub> emission rate (kg/kWh <sub>e</sub> )	0.830		

Table 3. KRW IGCC base case: energy consumption and CO<sub>2</sub> emissions.

Table 4. KRW IGCC with shift and Selexol: energy consumption and CO<sub>2</sub> emissions.

	Power CC (MW) Emis. (kg	sions
Coal mining	5.66 6,6	89
Limestone mining	0.59 7	72
Coal/limestone prep./sulfa	tor 2.60 12,2	38
IGCC with shift and Sele		23
Pipeline	21.20 21,2	40
Reservoir	0.0	4
TOTAL	83.73 74,8	66
Gross IGCC electric pow	er 403.4	
Net IGCC power at busb	ar 349.7	
Net electric power with e		
CO <sub>2</sub> emission rate (kg/k)		

#### REFERENCES

Condorelli, P., S. C. Smelser and G. J. McCleary (1991). Engineering and Economic Evaluation of  $CO_2$  Removal from Fossil-Fuel-Fired Power Plants, Volume 2: Coal Gasification-Combined-Cycle Power Plants. EPRI IE-7365, Electric Power Research Institute, Palo Alto, Calif.

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., et al., for the U.S. Department of Energy, Morgantown Energy Technology Center, Morgantown, W.Va.

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.

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, 1992).

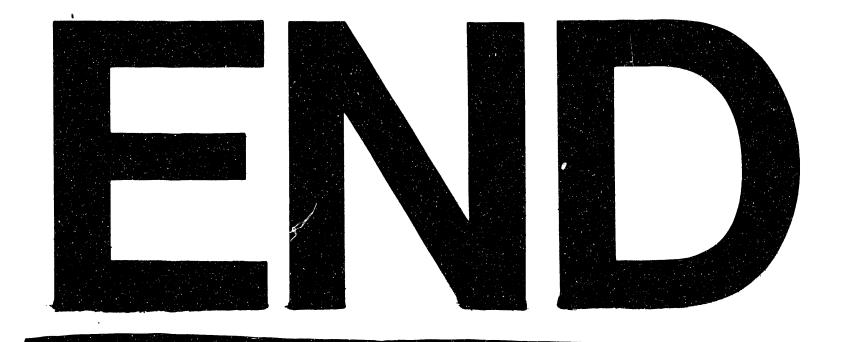
Marlund, G. (1983). Carbon dioxide emission rates for conventional and synthetic fuel. *Energy* <u>8</u>, 981-992.

Miller, W. R. (1985). Great Plains Coal Gasification Plant, Public Design Report. DOE/CH/10088-1874 (DE85016957). Morgantown Energy Technology Center, Grand Forks, N.D.

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, 1992).

#### DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.



# DATE FILMED 6/11/93

. and statements of