FOSSIL FUEL PROCESSING
TECHNICAL/PROFESSIONAL SERVICES

**ANALYSIS OF THERMAL
EFFICIENCY FOR HYDROGASIFICATION
AND GASIFICATION/METHANATION**

Prepared for

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COVER

Division of Fossil Fuel Processing
Contract
ET-78-C-01-3117

DECEMBER 21, 1979

A JOINT VENTURE OF
UOP INC. AND
SYSTEM DEVELOPMENT CORPORATION

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DRAFT

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EFFICIENCY FOR HYDROGASIFICATION
AND GASIFICATION/METHANATION

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SECTION 1 - SUMMARY

A Subtask of Task Order 019 (Rockwell Flash Hydrogasification) calls for a validation of the hypothesis that the thermodynamic efficiency of hydrogasification is greater than that of steam gasification and methanation. To confirm this potentially greater thermodynamic efficiency of hydrogasification, a study was made of carbon conversion and resulting thermal efficiency of methane production via hydrogasification as compared to gasification/methanation.

The alternative systems were first compared on an ideal process basis. This approach neglects limitations imposed by the thermodynamics and kinetics of the various chemical reactions and indicates maximum potential conversion of coal to methane. Results obtained from this ideal process comparison were then compared with similar data for commercial plant designs developed for each of the processes that take into account system inefficiencies and thermodynamic restraints.

SECTION 2 - CONCLUSIONS

A comparison of ideal processing sequences for hydrogasification and gasification/methanation confirms a potentially higher thermal efficiency for hydrogasification. Carbon conversions of 20 - 30% higher than given by gasification/methanation can ideally be achieved by hydrogasification, resulting in 20 - 30% higher thermal efficiencies. Unfortunately, the full extent of this advantage is not realized in commercial plant designs because of thermodynamic and kinetic limitations associated with direct carbon hydrogasification. The carbon-hydrogen-methane reaction equilibrium is not favorable to high methane concentrations at the high temperatures required for gasification of the reactant coal in a single stage process. This limitation is overcome by the use of a large excess of hydrogen, which favors increased carbon conversion within the required gasification temperature range but reduces thermal efficiency.

A comparison of both the cold gas (methane production only) and overall (methane plus byproducts) thermal efficiencies for the conceptual plant design developed by UOP/SDC for the Rockwell process with a similar plant design published for the Lurgi gasification/methanation process does, however, show about a 5% higher thermal efficiency for the Rockwell hydrogasification process. Because of the conceptual nature of the UOP/SDC Rockwell commercial plant design, this advantage may increase as more reliable reactor data are obtained and further plant optimization studies are performed.

SECTION 3 - DISCUSSION

3.1 IDEAL PROCESS COMPARISON

By considering only ideal processes for the conversion of coal to methane, based solely on stoichiometry, it is possible to compare conversion efficiencies of alternative processes without resorting to a comprehensive process design. Limitations imposed by thermodynamic equilibria and reaction kinetics are thus neglected, and a determination of maximum potential conversion efficiency is obtained.

In adapting this approach to the subject study, the following basic processing sequences were considered.

- 1) Gasification - consisting of:
 - a) Total gasification of coal with steam and oxygen followed by water gas shift to produce synthesis gas, and
 - b) Catalytic methanation of synthesis gas to produce methane.
- 2) Hydrogasification - consisting of:
 - a) Partial hydrogasification of coal to produce methane and a residual char (carbon) fraction, and
 - b) Gasification of char with steam and oxygen followed by water gas shift to produce hydrogen required for step (a).

Linden (1,2) presented the results of a similar analysis of gasification/methanation and hydrogasification assuming pure graphite as the reactant coal. A similar ideal approach to carbon processing in general was recently presented by Wojciechowski (4). In the present study, carbon conversion (lb carbon in methane/lb coal reactant carbon) and thermal efficiencies (heating value of methane product/heating value of reactant coal) were calculated assuming a reactant coal with a hydrogen to carbon ratio corresponding to Pittsburgh Seam

No. 8 coal. The basic equations for the above reaction steps used in this study are the same as those used by Linden and are considered representative for an ideal processing approach. Results of these calculations are given in Table 3.1 together with data for pure carbon taken from the Linden paper. Equations used in the analysis and a sample calculation for the Pittsburgh Seam No. 8 coal case are given in Appendix A.

Table 3.1 Comparison of Ideal Carbon Conversion and Thermal Efficiency of Gasification/Methanation vs. Hydrogasification

	<u>Gasification/Methanation</u>		<u>Hydrogasification</u>		Ratio*	
	<u>Reaction</u> <u>Carbon</u>	<u>Coal</u> <u>CH 0.84</u>	<u>Reaction</u> <u>Carbon</u>	<u>Coal</u> <u>CH 0.84</u>		
Carbon Conversion(1)	0.333	0.40	0.40	0.53	1.20	1.33
Cold Gas Efficiency (%) (2)	75.4		90.5		1.20	1.33

* Hydrogasification/Gasification

These data indicate that hydrogasification (including hydrogen production) is a potentially more efficient method of producing methane from coal than gasification/methanation. Assuming pure carbon as the reactant coal, the carbon conversion ratios for the ideal hydrogasification process is 0.40 as compared to a value of 0.333 for the ideal gasification/methanation process. Corresponding thermal efficiencies are 90.5% for hydrogasification and 75.4% for gasification/methanation. This represents an improvement of better than 20% in terms of carbon conversion and thermal efficiency. When considering a reactant coal with a hydrogen to carbon ratio equivalent to Pittsburgh Seam No. 8, the carbon conversion for the ideal hydrogasification process is 0.53 as compared to a value of 0.40 for the ideal gasification/methanation process. This represents more

(1) Lb Carbon in methane/lb carbon in coal

(2) Heating value of methane/Heating value of reactant coal

than a 30% improvement in terms of both carbon conversion and thermal efficiency (which is directly proportional to carbon conversion). However, as discussed below, much of this potential advantage for hydrogasification is lost in the transition from ideal to practical processing conditions.

3.2 PRACTICAL PROCESS COMPARISON

To determine the relative efficiencies of the two processes under practical processing conditions, the inputs to and expected outputs from two conceptual commercial plant designs were compared. One design was that developed by UOP/SDC for the Rockwell hydrogasification process; the other was of a gasification/methanation process based on Lurgi technology developed by C. F. Braun under the joint DOE-GRI coal gasification study program. Both plant designs are based on Pittsburgh Seam No. 8 coal feed with a nominal SNG production capacity of 250 billion Btu/day.

Table 3.2 summarizes the total plant energy inputs and outputs and resulting thermal efficiencies for the two processes. Both the cold gas and overall thermal efficiencies of the Rockwell hydrogasification process are greater than those of the Lurgi gasification/methanation process, although to a lesser degree than the ideal process comparison would indicate (Table 3.1). The Rockwell hydrogasification process produces about 1% more products in terms of heating value from about 4% less coal, indicating about 5% better overall thermal efficiency (63.4% as compared to 60.3% for the Lurgi process).

A further comparison of the two basic processing schemes is given in Table 3.3, which summarizes carbon conversion and thermal efficiencies on both a total plant coal feed basis and a net coal feed to the gasifiers basis. The ratio of the net gasifier coal feed to the total plant coal feed is a measure of the effectiveness of heat recovery and utilization within each process. Plant heat and power requirements that can not be satisfied by energy recovery within the process must be supplied by burning supplemental coal. For both designs, 84% of the total coal feed to the plant is required by the gasifiers; the remaining 16% is fed to the boilers for supplemental steam generation.

Process efficiency comparisons on a net coal to gasifier basis presented in Table 3.3 can be directly compared with ideal process efficiencies given in Table 3.1, since in both instances only coal that enters the conversion process is considered. On the basis of coal to gasifiers only, the carbon conversion for the Rockwell process is 0.40 lb/lb compared to 0.38 lb/lb for the Lurgi process. In terms of cold gas thermal efficiency (SNG production only), a value of 68.9% is obtained for the Rockwell process as compared to 65.6% for the Lurgi process. These data show that, although hydrogasification is still the more efficient process, the 0.38 lb/lb carbon conversion of the Lurgi gasification/methanation process is about 87% of the ideal process value of 0.40 lb/lb. A similar comparison for the Rockwell process indicates that the expected carbon conversion of 0.40 lb/lb is only 75% of the ideal value of 0.53 lb/lb. Since cold gas thermal efficiency is directly related to carbon conversion, the same result would be obtained when comparing cold gas thermal efficiencies.

Table 3.2 - Thermal Balance and Efficiency
Hydrogasification vs. Gasification/Methanation

	GASIFICATION/ (1) METHANATION		RATIO ROCKWELL/ LURGI
	LURGI PROCESS	HYDROGASIFICATION (2) ROCKWELL PROCESS	
Plant Input			
Coal			
Lb/Hr (10 ³)	1,524	1,470	
Btu/Day (10 ⁹)	453.7	437.4	0.96
Plant Output			
SNG - Btu/Day (10 ⁹)	249.8	253.8	1.02
By-Products (3)			
Btu/Day (10 ⁹)	<u>23.8</u>	<u>23.7</u>	
Total	273.6	277.5	1.01
Cold Gas Efficiency % (4)	55.1	58.0	1.05
Overall Thermal Efficiency % (5)	60.3	63.4	1.05

(1) C.F. Braun Data (Ref. 9)

(2) UOP/SDC Data (Ref. 7)

(3) By-Products include BTX/OIL, Ammonia and Sulfur

(4) (Btu/Day SNG)/(Btu/Day Total Plant Coal), %

(5) (Btu/Day Total Products Output)/(Btu/Day Total Plant Coal), %

Table 3.3 - Carbon Conversion and Thermal Efficiency
Hydrogasification vs. Gasification/Methanation

	GASIFICATION/(1) METHANATION		RATIO ROCKWELL/ LURGI
	LURGI PROCESS	ROCKWELL PROCESS	
Overall Plant			
Carbon Conversion - Lb/Lb(3)	0.319	0.335	1.05
Thermal Efficiency - %(4)			
Cold Gas	55.1	58.0	1.05
Overall	60.3	63.4	1.05
Gasifiers Only(5)			
Carbon Conversion - Lb/Lb(6)	0.380	0.400	1.05
Thermal Efficiency - %			
Cold Gas	65.6	68.9	1.05
Overall	71.8	75.6	1.05
Thermal Recovery Ratio(7)	0.84	0.84	1.0

(1) C. F. Braun Data (Ref. 5)

(2) UOP/SDC Data (Ref. 7)

(3) Lb carbon in methane/lb carbon in total plant coal

(4) See notes (4) and (5), Table 3.2

(5) Heating value of coal to gasifiers basis

(6) Lb carbon in methane/lb carbon in coal to gasifiers

(7) (Heating value of coal to gasifiers)/(Heating value of coal to plant)

One of the primary reasons for the reduction in practice in the thermal efficiency advantage of hydrogasification results from limitations imposed by the kinetics and thermodynamics of the primary reactions. Linden noted this fact in his paper on the conversion of solid fuels to high Btu gas when he stated that "Unfortunately, it is difficult to realize this basic advantage of hydrogasification because the methane-hydrogen-carbon equilibrium is not favorable to high methane concentrations under the most desirable operating conditions"(1). Data presented by Linden (Appendix B) indicate that high concentrations of methane can be obtained at relatively low temperatures and moderate pressures (1000 - 1200°F and 400 - 800 psia). This condition, however, results in low coal gasification rates, which would be impractical for commercial plant operation. This limitation is overcome by the use of a large excess of hydrogen, which favors increased carbon conversion to methane while controlling the large heat release associated with the highly exothermic hydrogasification reaction. Similar data supporting this fact are given by Lowry⁽⁶⁾ (Appendix C). Anthony and Howard⁽³⁾ also noted the favorable effect of hydrogen partial pressure on the kinetics of methane formation.

Unfortunately, this large amount of excess hydrogen must be separated from the product gas and recycled to the gasifier at the expense of additional power. In the Lurgi type gasification/methanation process, however, the gasification and synthesis steps are separated, and the optimum design conditions can be selected for each step. This factor also led IGT to the development of the two-stage Hygas process in which fed coal is first subjected to hydrogasification at a relatively low temperature followed by gasification of residual coal and char in a second, high-temperature stage⁽²⁾. Nevertheless, the basic superiority of the hydrogasification process is sufficient to overcome these thermodynamic and kinetic limitations, as demonstrated by the data in Table 3.3. It should be noted, however, that the data presented in Tables 3.2 and 3.3 for the Lurgi plant are based on a relatively proven process, whereas similar data for the Rockwell plant are based on conceptual designs and preliminary data. A further improvement in carbon conversion and thermal efficiency may be possible for the Rockwell process as more definitive design data are obtained and additional plant optimization studies are performed.

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APPENDIX A - SIMPLIFIED REACTIONS⁽¹⁾

A. GASIFICATION/METHANATION

1. Gasification

2. Shift

$$\text{Net CO after Shift} = (n + 0.25m) CO$$

$$\text{Net H}_2 \text{ after Shift} = (3n + 0.75m) H_2$$

3. Methanation

4. Combining steps 2 and 3

B. HYDROGASIFICATION (HYDROGEN BY CHAR STEAM/OXYGEN GASIFICATION)

1. Hydrogasification

2. Char Gasification

(1) Conversion of Solid Fossil Fuels to high Btu Pipeline Gas, H. R. Linden, Chem. Eng. Progress Symposium Ser. No. 54, 61, 76 (1965)

3. Shift

4. Combining Steps 2 and 3

SAMPLE CALCULATION

COAL: PITTSBURGH SEAM NO. 8

71.5 wt.% Carbon

5.02 wt.% Hydrogen

$$\text{H/C Ratio (Molal)} = \frac{(5.02)}{1} \bigg/ \frac{(71.5)}{12} = 0.84$$

$$C_n H_m = C_{1.0} H_{0.84}$$

A. GASIFICATION/METHANATION

1.0 GASIFICATION (EQ.A.1)

2.0 SHIFT/METHANATION (EQ.A.4)

CARBON CONVERSION = lb C in CH₄/lb C in coal

$$= (1.21 \times \frac{12 \text{ lb}}{\text{lb-mole}}) \bigg/ (3 \times \frac{12 \text{ lb}}{\text{lb-mole}})$$

$$= 0.40$$

B. HYDROGASIFICATION

(EQ. B.1)

$$\begin{aligned}\text{CARBON CONVERSION} &= (1.052 \times 12)/(2 \times 12) \\ &= 0.53\end{aligned}$$

Fig. 41.—Equilibrium Methane Concentrations for the Reaction $C + 2H_2 \rightleftharpoons CH_4$

APPENDIX C

COAL GASIFICATION

Percentage of methane in a mixture of methane and hydrogen in equilibrium with graphite and coke at various temperatures and 50 atm pressure.

(Ref. 6)