

PRODUCTION OF SNG BY FREE-FALL
DILUTE-PHASE HYDROGASIFICATION OF COAL

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INTRODUCTION

Hydrogasification of coal has been under investigation at PERC since the mid-1950's. Initial experiments were conducted with a 70-inch by 5/16-inch stainless steel, tubular reactor electrically heated to 800° C at 6000 psi and containing an 8-gram sample of 30 x 60 mesh coal (1).

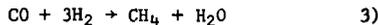
Experiments at PERC in the early 1960's were conducted using downward entrained flow, helical tube reactors 60 feet by 1/8-inch and 20 feet by 5/16-inch. Coal was entrained at a rate of 60 gm/hr in a 2 ft/sec hydrogen stream. Plugging problems due to particle agglomeration were encountered in the 500° to 550° C zone of the helical tube (2, 3).

These experiments led to development of the vertical free-fall, dilute-phase (FDP) reactor with large diameter so that coal particles were dispersed to reduce contact while in the plastic temperature range. Very rapid particle heating in the dilute-phase by mixing with concurrently fed hot hydrogen controlled agglomeration and eliminated the need for oxidative coal pretreatment. The FDP reactor was initially tested as the first stage of the two-stage HYDRANE process (4, 5). Experiments in the HYDRANE series used primarily hvAb coals with hydrogen/methane mixtures in the FDP reactor. However, a limited number of experiments were conducted which demonstrated that the FDP reactor alone could adequately convert lower rank coals and lignite in pure hydrogen. (6)

The basic objective of any gasification process to produce SNG is the conversion of coal, typically CH_{0.7} to methane, CH₄. Hydrogasification uses the approach of direct reaction of coal with hydrogen,



as opposed to formation of synthesis gas followed by methanation,



In the dilute-phase hydrogasification process, coal which has been washed, pulverized and dried is fed directly to the reactor without a requirement of oxidative pretreatment. Pretreatment, to destroy the coal's agglomerating property, may consume 9 pct of the volatile matter and 13 pct of the weight (7). A further advantage of hydrogasification is in the minimum use of the methanation reaction shown in Equation 3. This reaction is highly exothermic, but the heat cannot be used directly in the gasifier because the temperature must be limited to 450° C for protection of catalysts. In the DPH process, typically 65-75 pct of total methane product may be produced directly in the FDP reactor.

A block diagram of the DPH process is shown in Figure 1. Raw pulverized coal and heated hydrogen were fed to the reactor and char and product gas were recovered as products. Char from the reactor may be used either in hydrogen generation or as a fuel for power and steam generation.

EXPERIMENTAL FACILITY

A schematic of the laboratory experimental facility is shown in Figure 2. Coal pulverized to 80 pct minus 200 mesh (U. S. standard sieve series) was initially loaded into a ground-level, low-pressure charging hopper and transferred in the dense phase by nitrogen to the first high-pressure lock hopper. Pressure was equalized between lock hoppers and coal transferred between them by gravity flow. Each lock hopper was 10-inch diameter schedule 120 carbon steel with a stainless steel liner and held approximately a 100 lb coal capacity.

Coal was fed from the second lock hopper by a rotary vane feeder through a water-cooled nozzle, a 0.3-inch tube, to the reactor at rates from 9 to 47 lb/hr. The reactor consisted of an electrically heated 304 stainless steel pipe, 3.26-inch internal diameter and enclosed in a 10-inch carbon steel pressure vessel. Reactor lengths of 5 and 9 feet were used. Hydrogen gas was heated by passing through a helical coil of tubing located in the annulus between the hot reactor wall and pressure vessel. It was injected at the reactor head concurrently downward with the coal. Char and product gas were separated in a disengaging zone below the reactor.

Product gas samples were automatically analyzed by an on-line gas chromatograph at 15 minute intervals. All experimental data, including gas analyses, were stored on a PDP-11 computer.

Char was collected at the base of the facility in two air-cooled, stainless steel receivers which were alternately filled and emptied during a test. Use of dual feed lock hoppers and char receivers allowed continuous operation.

EXPERIMENTAL PROGRAM AND RESULTS

In the present single-stage FDP reactor it was anticipated that only the more reactive lower rank coals would have adequate carbon conversion for SNG production. Therefore, only Illinois #6 hvCb coal and North Dakota lignite have been tested for the DPH process. Objectives of the experimental program were to demonstrate feasibility and operability of the FDP reactor for SNG production through both long and short duration parametric experiments. Data were obtained on yield and distribution of hydrogenation products to determine optimum test conditions and provide a thorough design data base for scale-up to a larger process development unit. Parameters in the test program were coal type, reactor length, hydrogen/coal ratio and reactor throughput.

Results of several experiments are summarized in Table 1 and typical analyses of coal and lignite are presented in Table 2. All tests were conducted at 1000 psig, with the reactor wall at 900° C. No thermocouples were located internally below the coal injection point to eliminate any potential blockage. Feed gas in all experiments was over 99 pct hydrogen. Both coal and lignite were pulverized and screened to 80 pct minus 200 mesh (all minus 100 mesh). Average particle size for lignite and coal was 73.4 and 82.6 μm respectively, determined by screen analysis. Conversion was calculated on the basis of ultimate analysis and actual feed and recovery weights of coal and char, with no forcing to 100 pct carbon or ash balance.

Experiments #124 and #128 were typical of those conducted with Illinois #6 coal using the five-foot heated reactor. Test times were limited by the single coal hopper that was used prior to installation of the dual lock hopper feed system. These experiments represent a 50 pct variation in coal feed rate, with two hydrogen/coal ratios tested at each feed rate. In both experiments carbon conversion and methane yield varied directly with the hydrogen/coal ratio. Char particles from these experiments showed an average diameter of 588 μm by screen analysis, with size independent of test conditions. Neither experiment produced a carbon conversion necessary for balanced plant operation, indicating the necessity for a longer

residence time in the reactor.

Major facility modifications following tests with Illinois #6 coal (dual lock hoppers) permitted extended continuous operation. The five-foot reactor was replaced with a nine-foot reactor having heated lengths of one to nine feet in two-foot intervals. Testing then resumed using North Dakota lignite.

High moisture and oxygen contents of the lignite led to production of more water and carbon oxides than with hvCb coal. The higher CO content in the product gas stream reduced total methane to typically 75 pct with lignite as compared to 90 pct with bituminous coal.

In experiment #134, the first five-foot section of the reactor was operated at 900° C while the lower four-foot section was only heated to 300° C. This was to minimize the possibility of moisture condensation and char packing by permitting water to be removed through the product gas system. Following this procedure the experiment was conducted for a period of 45 hours at an average lignite feed rate of 12.5 lb/hr. No reactor problems were encountered; however the test was terminated by feeder stoppage due to fine particulates packing around the shaft, causing it to seize. Balanced plant operation was achieved with 44 pct carbon conversion, but product gas hydrogen-to-carbon monoxide ratio was higher than desired for final methanation.

The complete reactor was operated at 900° C in experiment #135 in order to achieve high carbon conversion at low hydrogen/coal ratios. Two hydrogen/coal ratios were tested, resulting in carbon conversion of 44 and 50 pct with low hydrogen/carbon monoxide ratios in product gas. Ratios of 3.36 and 2.8 were obtained as compared to ratios in excess of 7.0 in previous experiments. These values are consistent with requirements for final cleanup methanation with no residual hydrogen separation. Product gases from 135A and 135B had calculated heating values of 939 and 1008 Btu/scf, assuming CO methanation.

Experiment #136 was conducted for 22.6 hours at conditions nearly duplicating test #135B to verify results at these conditions. Figure 3 is typical of methane and hydrogen composition of the product gas for the duration of this test.

Experiment #137 was conducted to determine the effect of throughput on FDP reactor performance. Coal feed rate was varied from 15.9 to 47.0 lb/hr corresponding to a throughput range of 276 to 816 lb/ft²hr. Feed gas rates were varied proportionally to maintain nearly constant hydrogen/coal ratio. Conversion varied inversely with throughput, indicating a requirement for increased reactor length at high throughput conditions. However, under all test conditions, steady state reactor operation was easily maintained and carbon conversion to methane remained nearly 62.5 pct. No oil formation was detected in either char receivers or liquids traps at any operating conditions.

In experiment #139 data was obtained on the effect of hydrogen/coal variation upon carbon conversion and gas composition at a nominal 25 lb/hr lignite feed rate. A 68 pct change in hydrogen/coal ratio was tested, resulting with a 23 pct increase in methane yield per pound of coal. All other changes were relatively small.

CONCLUSIONS

Dilute-phase hydrogasification has been demonstrated in successful, continuous, long duration experiments. Carbon conversion necessary for balanced plant operation with lignite has been demonstrated at hydrogen/coal ratios producing a high-Btu SNG with no residual hydrogen separation requirement. High carbon selectivity to gas phase products has been demonstrated with no benzene and only trace oil formation. Parametric testing has established effects of throughput upon carbon conversion and product distribution. Steady operation was achieved at a throughput of over

800 lb/ft²hr. Reproducibility of test results has been demonstrated by duplicated test operation.

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TABLE 1.-Dilute-phase hydrogasification of coal

Experiment No.	124A	124B	128A	128B	134	135A	135B
Coal Type	Illinois	Illinois	Illinois	Illinois	Lignite	Lignite	Lignite
Time (hr)	4.5	1.43	3.75	4.17	45.0	4.2	8.5
Reactor length (ft).	5	5	5	5	5	9	9
Temperature (° C) ..	900	900	900	900	900	900	900
Pressure (psig) ..a	1000	1000	1000	1000	1000	1000	1000
Coal rate (lb/hr) ..	15.0	15.0	10.03	10.03	12.4	14.8	14.8
Throughput (lb/ft ² hr) ^a	260	260	174	174	215	257	257
Feed gas/coal (scf/lb) ^a	9.31	7.93	13.78	12.10	15.6	17.0	10.6
Product gas/coal (scf/lb) ^a	9.75	8.03	14.53	12.17	16.8	18.8	12.3
Product gas comp. (vol%)							
H ₂	42.1	32.5	47.4	40.1	54.3	48.0	41.4
CH ₄	48.9	58.4	46.1	52.1	30.6	30.5	33.3
C ₂ H ₆	1.5	0.4	-	-	-	-	-
CO	5.3	6.2	5.7	6.3	7.9	14.3	14.8
CO ₂	2.0	1.9	0.4	0.6	6.5	6.6	10.0
H ₂ S	-	-	-	.4	0.3	0.2	0.2
N ₂	0.2	0.2	0.3	.3	.3	.3	.3
Conversion (wt%) ^b							
Carbon	31.6	27.2	36.0	29.8	44.2	44.0	50.7
Hydrogen	76.2	72.5	80.8	77.8	83.5	83.3	88.4
Maf coal	40.6	37.0	45.1	39.7	58.1	57.6	63.7
Feed hydrogen consumption (scf/lb) ^a	5.09	4.47	6.84	7.67	6.48	7.98	5.51
Product yield/coal fed							
Methane (scf/lb) ^c	5.28	5.19	7.53	7.11	6.47	8.42	5.92
Char (lb/lb) ^d	0.64	0.67	0.60	0.64	0.46	0.47	0.40
Water (lb/lb) ^e04	.04	.03	.03	.13	.11	.11
Carbon recovery (wt%)	94.4	97.5	97.8	102.0	98.0	100.5	95.0
Heating value (Btu/scf) ^d	803	892	755	852	705	939	1008
CH ₄ made in reactor (%)	90.2	90.4	89.0	89.2	79.5	68.1	69.2

TABLE 1-cont.

Experiment No.	136	137-A	137-B	137-C	137-D	139-A	139-B
Coal type	Lignite						
Time (hr)	22.6	12.43	12.51	2.19	10.71	7.56	8.01
Reactor length (ft) ..	9	9	9	9	9	9	9
Temperature (°C)	900	900	900	900	900	900	900
Pressure (psig)	1000	1000	1000	1000	1000	1000	1000
Coal rate (lb/hr) ^a ..	14.6	15.90	26.29	47.31	14.48	26.11	24.64
Throughput (lb/ft ² hr) ^a	233	274	454	816	250	453	428
Feed gas/coal (scf/lb) ^a	9.91	9.59	9.76	11.23	12.13	8.90	14.94
Product gas/goal (scf/lb) ^a	12.3	12.64	12.53	11.86	14.03	11.78	16.07
Product gas comp. (vol%)							
H ₂	46.7	45.6	49.5	56.8	53.8	44.9	51.8
CH ₄	34.5	34.0	31.1	26.4	30.6	33.5	32.3
C ₂ H ₆	-	0.1	0.1	0.2	-	-	-
CO	11.2	11.6	11.2	9.0	10.3	14.9	12.2
CO ₂	7.1	8.2	7.5	7.0	4.9	6.1	3.1
H ₂ S	0.2	.2	.3	.3	.1	.4	.2
N ₂3	0.3	0.3	0.3	0.3	0.2	0.3
Conversion (wt%) ^b ..							
Carbon	48.2	44.1	39.7	30.6	47.1	38.9	40.4
Hydrogen	82.8	83.9	81.3	75.3	80.6	82.3	85.8
Maf coal	56.0	59.3	55.7	46.6	59.9	54.3	55.8
Feed hydrogen comp. (scf/lb) ^a	4.17	3.83	3.56	4.49	4.58	3.61	6.61
Product yield/coal fed							
Methane (scf/lb) ^d ..	5.62	5.76	5.30	4.20	5.71	5.80	7.14
Char (lb/lb) ⁵	0.44	0.46	0.49	0.58	0.44	0.49	0.48
Water (lb/lb) ⁵13	.13	.20	.28	.14	.08	.12
Carbon recovery (wt%)	95.0	93.1	93.8	99.0	94.8	94.3	99.1
Heating value (Btu/scf) ^d	856	875	823	694	760	1005	834
CH ₄ made in reactor (%)	75.5	74.6	73.5	74.6	74.8	69.2	72.5

^aAs received ^bMoisture free ^cPrior to methanation ^dCalculated assuming methanation

Table 2. Typical coal and lignite analysis

	<u>Illinois #6 hvCb Coal</u>	<u>North Dakota Lignite</u>
<u>Proximate (wt pct)</u>		
Moisture	1.0	9.7
Volatile matter	35.2	38.2
Fixed carbon	53.7	42.5
Ash	10.1	9.6
<u>Ultimate (wt pct dry)</u>		
Hydrogen	4.8	4.9
Carbon	71.8	56.9
Nitrogen	1.7	0.7
Oxygen (by difference)	10.1	25.7
Sulfur	1.4	1.2
Ash	<u>10.2</u>	<u>10.6</u>
	100.0	100.0

hvCb from Orient #3 mine, Freeman Coal Co., Waltonville, IL.
Lignite from Beulah seam, North Dakota.

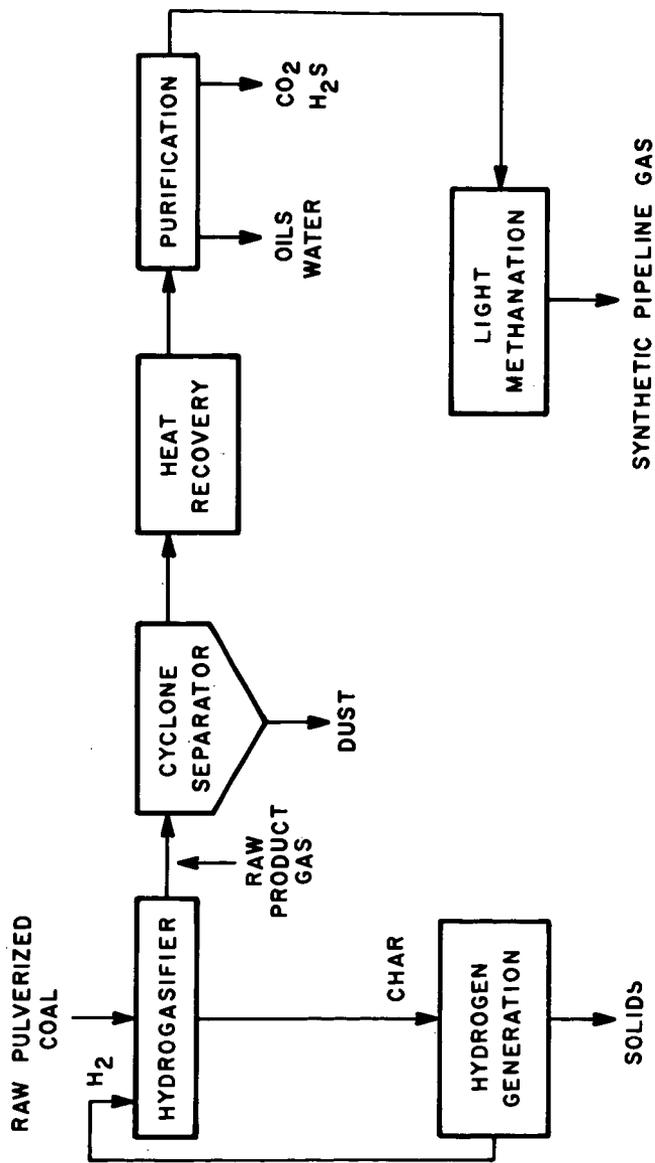


Figure 1 - Dilute phase hydrogasification process

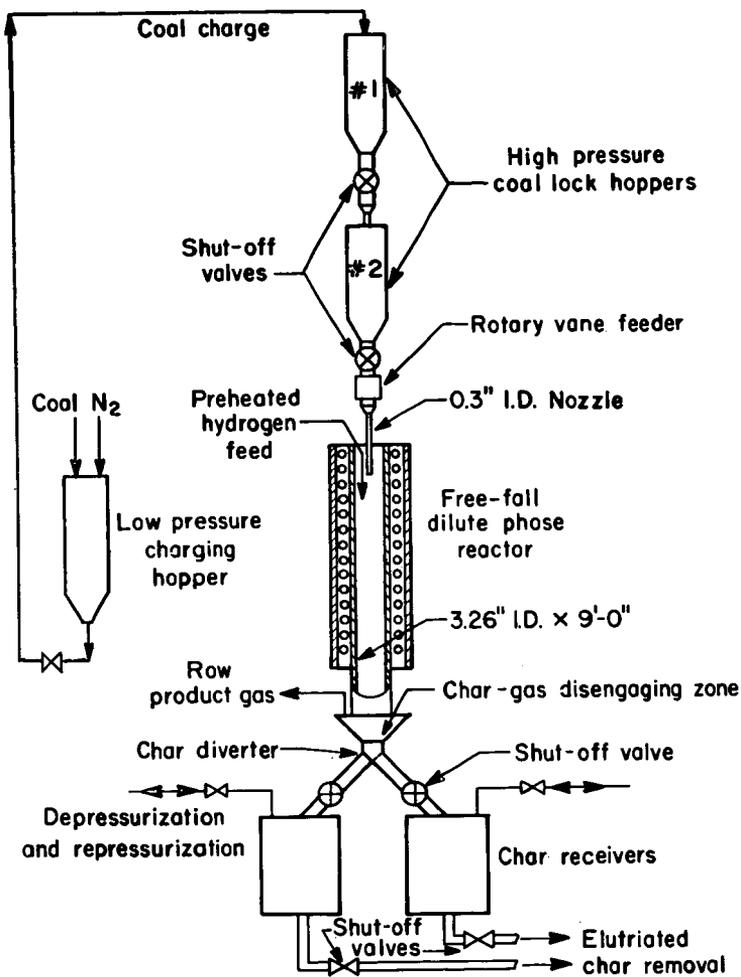


Figure 2- Dilute-phase hydrogasifier

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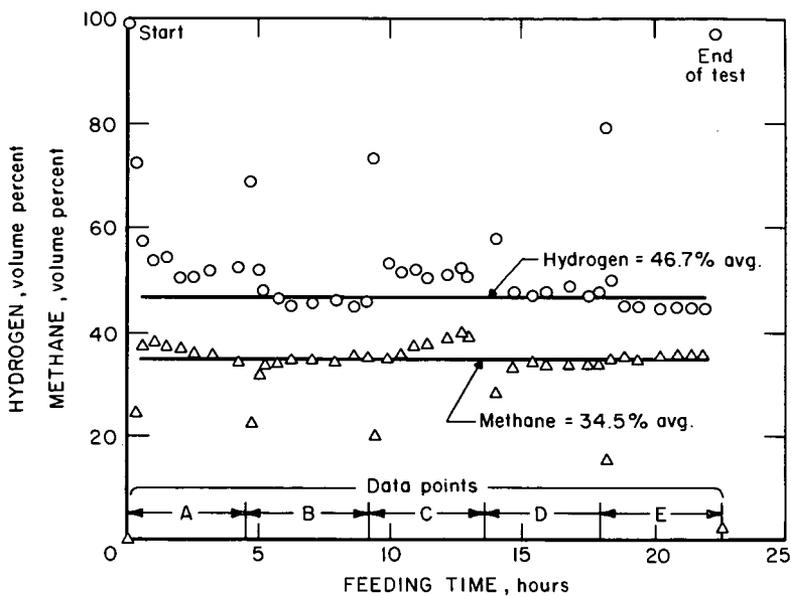


Figure 3 - Product gas composition vs time experiment #136
 lignite feed, FDP reactor T=900°C P=1000 psig
 L=9ft.