

NOT FOR PUBLICATION

Presented Before the Division of Gas and Fuel Chemistry  
American Chemical Society  
Atlantic City, New Jersey, Meeting, September 13-18, 1959

PRODUCTION OF PIPELINE GAS BY HIGH-PRESSURE,  
FLUID-BED HYDROGASIFICATION OF CHAR

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Production of high-methane-content (pipeline) gas by direct hydrogenation of coal or low-temperature char<sup>2, 9, 14, 15</sup> has three major advantages over the two-step partial coal oxidation-synthesis gas methanation process:<sup>1, 13</sup> large reduction, or potential elimination, of oxygen requirements; elimination of the extreme synthesis gas purification requirements prior to catalytic methanation; and greater thermal efficiency through reduction of exothermic heats of reaction. Complete feed utilization will result if less than half of the more reactive coal or char constituents is hydrogasified and the residue is used as a source of hydrogen. However, essentially complete hydrogasification of lignites and some subbituminous coals may be feasible; hydrogen could then be produced by the reforming of a portion of the product gas or the primary natural gas supply.

In earlier phases of this investigation,<sup>5, 14</sup> it was demonstrated in batch reactor tests that gases containing 60-80 volume % methane could be produced by hydrogasification of the lower-rank coals at 1350°F. and 2500-4000 p.s.i.g. By adjustment of hydrogen-coal ratios, gasifications on a moisture-, ash-free (MAF) basis up to 80 wt. % were obtained with bituminous coal, and over 90 wt. % with lignite. However, the residues from bituminous coal hydrogasification were severely agglomerated. As this would hinder smooth operation of a continuous hydrogasification reactor, a pretreatment step yielding nonagglomerating, reactive chars was needed. Optimum reactivity and substantial reduction of agglomeration tendencies of bituminous coal were obtained by fluidized pretreatment in air and nitrogen for about one hour at a maximum temperature of about 600°F; pretreatment with steam as the fluidizing medium did not reduce agglomeration tendencies to the same extent, and pretreatment with carbon dioxide produced chars with the highest agglomeration tendency and also lower reactivity.<sup>5</sup> Lignite, although essentially nonagglomerating during hydrogasification, was benefited by elimination of CO<sub>2</sub>; reactivity was only slightly increased by fluidized pretreatment at about 500°F.<sup>5</sup>

REACTOR DESIGN

After the feasibility of preparing sufficiently reactive, non-agglomerating feeds had been established, a design for a fluid-bed hydrogasification unit was developed. To obtain reliable information on the effects of the process variables, it was deemed necessary to have positive control over bed depth. This required parallel upward flow of pulverized coal or char, and of hydrogen, with discharge through a standpipe. It was felt that if a single-stage cocurrent-flow reactor could not produce gas of the desired heating value, or give hydrogasification yields required for a balanced process, multistage operation could be simulated by a series of operations in a single reactor.

The main design problem was selection of the bed depth, hydrogen superficial velocity and coal feed rate which, at nominal design operating conditions of 1400°F., 1000 p.s.i.g. and 60-325 mesh particle size range, would give a) sufficient agitation for free movement of the bed, b) sufficient gas residence time for utilization of most of the hydrogen feed, and c) hydrogen/coal ratios sufficient for gasification of a substantial portion of the coal feed. Tests were made in glass models of upflow reactors with carbon dioxide at a atmospheric pressure and temperature to simulate the mass velocity of hydrogen at reaction conditions. In a 2-inch diameter reactor with a 1-inch standpipe, adequate fluidization of a 5-foot char bed was obtained at a superficial gas velocity of about 0.06 ft./sec. This corresponded to a gas residence time of 1.3-1.4 seconds and, at a char feed rate of about 4 lb./hr., to a char residence time of 30 minutes and an equivalent hydrogen/char ratio of 20 SCF/lb. It appeared from the batch reactor test results that these conditions should give acceptable gasification results; however, the reactor was designed to accommodate an approximately 9-foot bed for greater operational flexibility.

#### APPARATUS

The high-pressure semicontinuous coal hydrogasification pilot unit consists of an interconnected pressure vessel assembly that includes a reactor, a char feed hopper, a screw feeder and an ungasified-char-residue receiver (Figure 1). The reactor is designed for a working pressure of 3500 p.s.i.g. at 1500°F. and is fabricated of 19-9 DL alloy. It has an inside effective length of 113 inches, an outside diameter of 5 inches, and an inside diameter of 2 inches. A 1-inch diameter standpipe, of a selected height, provides the reaction annulus, controls the height of the char bed, and serves for the removal of the residual char and product gases from the top of the reactor. Self-sealing closures at the top and bottom of the reactor are of the modified Bridgman type with a stainless steel seal ring.

The char feed hopper is fabricated of Type 316 stainless steel and has an inside diameter of 5 inches, an outside diameter of 6-3/4 inches, and has recently been extended to an inside height of 120 inches. It is fitted with confined gasket-type closures at the top and the bottom. Capacity of the hopper is about 40 pounds of char. In most of the tests reported here, the original hopper with an inside height of 60 inches and half of the present char capacity was used. The hopper is parallel to the reactor and is joined to it by a horizontal feed-screw-housing 24 inches in length (see Figure 2 for details).

The ungasified-char receiver has the same dimensions as the original hopper, and is also fabricated of Type 316 stainless steel. The top of the residue receiver is joined to the bottom of the reactor by a short tube with a self-sealing closure, (modified and full Bridgman, respectively). The residue receiver also acts as a separator for the product gas and the ungasified char. The product gas is passed through a porous stainless steel filter, a water-cooled condenser, and a bank of gas filters for final cleanup. Pressure on the hydrogasification unit is maintained by an externally loaded back-pressure regulator.

Char is transferred through the housing connecting the feed hopper and the reactor by means of a rotating spiral screw with an outside diameter of 5/8 inch, a root diameter of 1/4 inch, and a pitch of 0.40 inch. Delivery of char to the screw within the feed hopper is by gravity. A variable-speed electric motor drive, with belt coupling, rotates the screw. Normal operation is at 25-85 r.p.m. for char rates of 1.5 to 6 lb./hr. Commercial electrolytic hydrogen, recom-

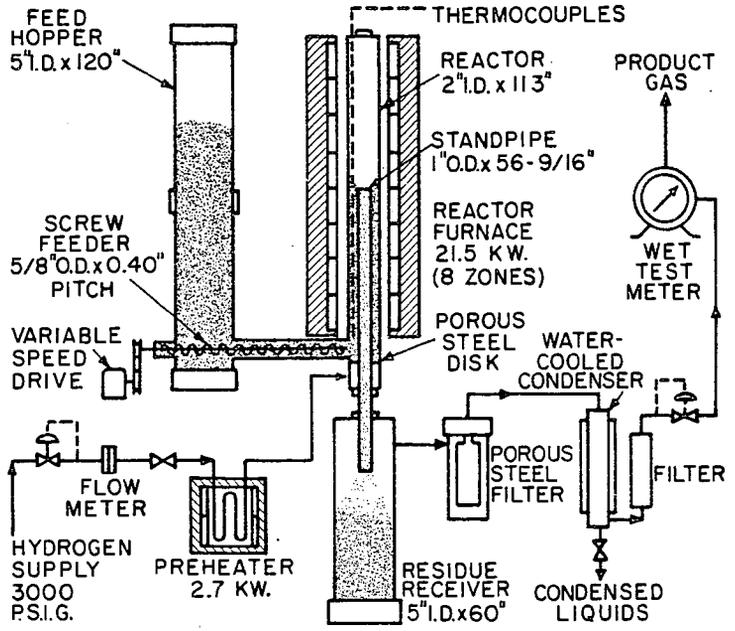


Fig. 1.-COAL HYDROGASIFICATION PILOT UNIT

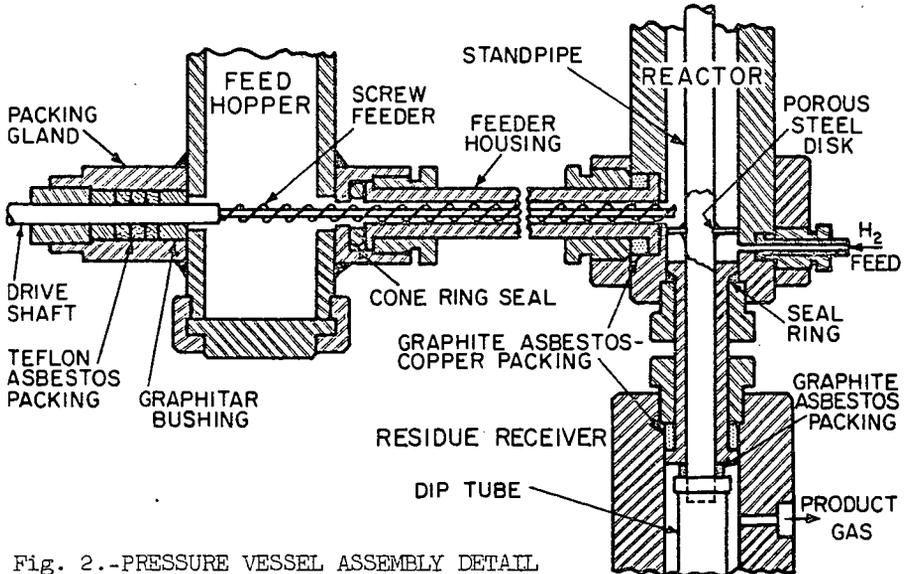


Fig. 2.-PRESSURE VESSEL ASSEMBLY DETAIL

pressed to 3000 p.s.i.g., is fed from a manifolded cylinder bank. A regulator reduces the pressure to a level depending on the desired reactor pressure and hydrogen flow rate. Flow is controlled by a manually operated needle valve, and is metered by a plate orifice with flange pressure taps. Hydrogen enters the reactor preheated to 150°F.

The char bed (reaction) volume is 0.07391 cu.ft. when a nominal 5-foot standpipe (4.714-foot true height) of 1-inch diameter is used. The reactor cross-sectional area, based on a 2-inch by 1-inch annulus, and corrected for two 1/4-inch diameter thermowells that extend the length of the reactor, is 0.01568 sq.ft. A sintered steel disk, of 0.0025-inch mean pore size, which is fastened to the standpipe just below the point where char from the screw feeder enters the reactor, serves as a base for the char bed and as a distributor for the feed hydrogen as it enters the reactor below the disk.

The reactor is heated externally by a 21.5 kw. electric furnace having eight individually controlled heating zones. Char bed temperatures are sensed in ten locations with chromel-alumel thermocouples. Feed hydrogen and gas stream temperatures are also sensed with chromel-alumel thermocouples.

Reactor pressure, differential pressure across the reactor and differential pressure across the hydrogen orifice are continuously recorded. Product gas volume is measured with a wet test meter, and product gas specific gravity is indicated and recorded by a continuous gravitometer.

#### EXPERIMENTAL PROCEDURE

Feed batches were prepared by crushing and screening the char to a 60-325 mesh size, and drying in air at 110°C. in a forced-convection oven. After the char had been charged to the hopper, the preheated unit was purged with nitrogen and pressurized with hydrogen. The hydrogen orifice meter was then calibrated against the wet test gas meter, the desired hydrogen feed rate established, and the char screw feeder started at a preset rate.

When the char bed reached the top of the standpipe, the differential pressure across the reactor became constant. This differential was normally 20-25 inches of water column, which agreed closely with the value predicted from the bulk density of the expanded char bed; severe fluctuations of differential pressure were an indication of mechanical operating difficulties, such as bypassing of feed hydrogen around the porous steel distributing disk, or plugging of the reactor at the char feed inlet.

The steady-state operating period began when the product gas specific gravity reached a constant value. A composite gas sample was continuously bled into a 10 cu.ft. water-sealed gas holder during the steady-state period; spot gas samples were also taken periodically to confirm other observations relating to attainment of steady operating conditions.

Upon termination of the test, which normally lasted from 4-1/2 to 5-1/2 hours, the unit was depressurized, purged and allowed to cool. Char remaining in the feed hopper, solid residues and condensed liquids were then removed and weighed. Samples of char feed and solid residues were subjected to proximate, ultimate and sieve analyses, and their heating values were determined. Product gases were analyzed with a Consolidated Engineering Co. Model 21-103 mass spectrometer. Carbon monoxide content was determined by infrared

analysis with a Perkin-Elmer Model 12-C infrared spectrometer.

The properties of the composite gas samples, and the char and residue weights corrected to a dry basis, were used in the computation of hydrogasification test results. The char feed rate was computed from the difference between the charged and recovered weights, and from the total feed time. Hydrogen and product gas volumes for the steady-state period were corrected to 60°F. and 30 inches of mercury pressure, and are reported as standard cubic feet (SCF) on a dry basis. Gas heating values and specific gravities were computed from the gas analysis; heating values are reported at 60°F., 30 inches of mercury pressure and saturated with water vapor, whereas specific gravities are for dry gas referred to dry air.

## RESULTS

A low-temperature bituminous coal char (Montour No. 10 Mine) supplied by the Research and Development Division of the Consolidation Coal Co. was used in this study. This material was produced in a fluidized carbonization process at much more severe conditions than those employed in the earlier pretreatment study; in batch hydrogasification tests it produced a free-flowing residue, but had significantly less reactivity than chars produced at optimum pretreatment conditions.<sup>5</sup> It was selected for a study of process variables because of its ready availability and uniform properties, and the prospect of minimum handling difficulties. Unlike the feed material employed in the laboratory semiflow study reported by Hiteshue and others,<sup>12</sup> the char was not impregnated with a catalyst. Typical properties of the feed char and the hydrogasification residues are given in Table 1, and selected results for operation at 500, 1000, 1500 and 2000 p.s.i.g., 1400° and 1500°F. nominal temperature and nominal char feed rates of 2, 4.5 and 6 lb./hr., are given in Table 2. Precise control of char feed rate was not practical; actual values for each feed rate level varied considerably, and the overall range was 1.6-6.3 lb./hr. Normally, hydrogen feed rates corresponded to a superficial velocity of about 0.06 ft./sec., the level required to maintain free flow of char through the reactor; this was equivalent to rates ranging from 34-35 SCF (dry)/hr. at 500 p.s.i.g., to 130-135 SCF (dry)/hr. at 2000 p.s.i.g. Consequently, with the exception of a few tests at a higher hydrogen rate than the minimum, the hydrogen residence time based on the free reaction volume with the 5-foot stand-pipe was nearly constant at 1.2-1.5 minutes.

The average reactor temperatures reported in Table 2 are based on measurements taken 1, 8, 18, 28, 42, and 54 inches above the bottom of the char bed. The closer spacing at the bottom, where temperatures are lowest, is partially responsible for the substantial difference between the average and maximum temperatures; the latter closely approach the nominal temperature levels of 1400° and 1500°F.

The effects of char feed rate and pressure on hydrogasification results at 1400°F. are shown graphically in Figure 3. The char feed rate expressed as space velocity or reciprocal space velocity is used as the independent variable. Char residence times can be computed by assuming a typical bulk density of the expanded bed of 25 lb./cu.ft. The available data for the low-temperature bituminous coal char exhibit the expected trends: increase in the percent of char hydrogasified (on a moisture-, ash-free basis) with increases in pressure and decreases in char space velocity; increase in gaseous hydrocarbon

Table 1.-PROXIMATE, ULTIMATE AND SCREEN ANALYSES OF CHAR FEEDS AND RESIDUES

Run No.	19		17		45		13		43	
Operating conditions										
Reactor pressure, p.s.i.g.	521		515		1007		1031		1015	
Maximum reactor temperature, °F.	1415		1405		1405		1410		1510	
Char feed rate, lb./hr. <sup>a</sup>	2.07		5.95		1.92		5.26		1.97	
Sample	Feed	Residue								
Proximate analysis, wt. %										
Moisture	0.1	7.1	0.4	4.3	0.0	6.2	0.1	7.0	0.0	6.4
Volatile matter	17.2	3.2	17.1	3.6	17.4	3.8	17.6	4.2	17.1	2.2
Fixed carbon	74.7	79.9	74.4	82.7	75.1	78.7	74.2	79.9	75.3	79.1
Ash	8.0	9.8	8.1	9.4	7.5	11.3	8.1	8.9	7.6	12.3
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0
Ultimate analysis, wt. % (dry basis)										
Carbon	77.6	85.6	77.4	84.7	78.7	83.9	77.6	84.4	78.1	83.0
Hydrogen	5.15	1.55	3.32	1.82	3.13	1.57	3.30	1.82	3.18	1.28
Sulfur	0.93	0.65	0.93	0.89	0.93	0.76	1.05	0.97	0.93	0.67
Ash	8.0	10.6	8.1	9.8	7.5	12.0	8.1	9.6	7.6	13.2
Nitrogen + oxygen (by difference)	10.32	3.30	10.25	2.70	9.68	1.77	9.87	3.21	10.10	1.85
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0
Heating value, B.t.u./lb. (dry basis)	12,877	13,035	12,765	13,214	12,889	12,859	12,133	13,256	12,743	12,584
Screen analysis, wt. % <sup>b</sup>										
+ 40 mesh	0.0	0.0	0.0	0.0	0.1	0.4	0.0	0.0	0.2	0.2
+ 60 mesh	0.0	0.4	0.0	0.2	0.4	0.2	0.2	0.0	0.2	0.4
+ 80 mesh	5.2	1.2	7.2	2.4	10.1	2.2	2.8	1.2	9.4	1.0
+100 mesh	22.0	17.8	21.6	19.6	10.6	7.6	14.2	10.2	12.6	5.6
+140 mesh	10.0	11.2	3.4	8.2	18.4	15.4	9.2	8.6	18.6	14.6
+200 mesh	33.0	33.0	34.6	38.8	23.8	20.0	31.6	32.0	22.2	20.0
+325 mesh	20.6	26.0	19.2	37.6	27.2	29.6	24.4	35.0	25.7	31.8
-325 mesh	9.2	10.4	9.0	14.2	9.4	24.6	17.6	15.0	11.1	26.4
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0

Run No.	44		28		31		40		32	
Operating conditions										
Reactor pressure, p.s.i.g.	1001		1518		1512		2012		2004	
Maximum reactor temperature, °F.	1505		1405		1435		1425		1415	
Char feed rate, lb./hr. <sup>a</sup>	6.08		1.76		5.27		2.03		5.75	
Sample	Feed	Residue								
Proximate analysis, wt. %										
Moisture	0.0	5.5	0.1	11.0	0.1	6.9	0.0	10.0	0.1	5.6
Volatile matter	17.9	2.0	16.8	2.9	17.6	2.8	19.7	4.4	19.7	4.1
Fixed carbon	74.1	82.3	76.4	76.0	75.7	81.7	73.1	74.2	73.0	79.1
Ash	8.0	10.2	6.7	10.1	6.6	8.6	7.2	11.4	7.2	11.2
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0
Ultimate analysis, wt. % (dry basis)										
Carbon	78.4	85.5	78.8	83.1	78.8	85.6	78.5	82.7	78.5	83.4
Hydrogen	3.19	1.25	3.16	1.60	3.16	1.68	3.22	1.73	3.22	2.20
Sulfur	0.95	0.67	1.06	1.27	0.98	1.15	0.93	0.78	0.93	0.93
Ash	8.0	10.8	6.7	11.3	6.6	9.2	7.2	12.7	7.2	11.9
Nitrogen + oxygen (by difference)	9.96	1.78	10.28	2.73	10.46	2.39	10.15	2.09	10.15	1.57
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0
Heating value, B.t.u./lb. (dry basis)	12,760	12,966	13,015	12,892	13,032	13,452	13,011	12,432	13,011	13,142
Screen analysis, wt. % <sup>b</sup>										
+ 40 mesh	0.0	0.2	0.0	0.0	0.0	0.9	0.0	0.2	0.2	0.2
+ 60 mesh	0.4	0.2	0.0	0.0	2.0	1.0	1.4	0.2	1.8	1.0
+ 80 mesh	9.2	2.8	24.8	5.2	18.4	7.5	9.0	3.0	13.2	6.3
+100 mesh	11.4	10.4	19.1	11.2	15.0	15.0	12.8	5.8	12.7	12.0
+140 mesh	19.2	20.8	21.0	23.9	20.9	24.9	11.6	13.8	20.0	19.1
+200 mesh	22.0	23.4	20.7	25.7	18.4	22.1	25.2	18.4	21.7	20.9
+325 mesh	26.8	27.3	11.7	19.0	16.2	17.5	32.4	27.3	19.7	21.0
-325 mesh	11.0	14.4	3.7	17.0	9.1	11.1	7.6	35.8	10.7	19.5
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0

a. Based on weight of dry char.  
 b. U. S. standard sieve series.

Table 2.-OPERATING DATA FOR FLUID-BED HYDROGASIFICATION OF LOW-TEMPERATURE CHAR AT HIGH PRESSURES

Feed Char: Low-Temperature Bituminous Coal Char, Consolidation Coal Co. Montour No. 10 Mine.<sup>a</sup>  
 Particle Size: 60-325 Mesh, U. S. Standard Sieve Series. Char Bed Height: 4.714 ft. Char Bed Volume: 0.07391 cu.ft.

Run No.	19	17	45	11	13	43	22	44	28	25	31	40	41	32
Operating conditions														
Reactor pressure, p.s.i.g.	521	516	1007	1046	1031	1015	1015	1001	1518	1527	1512	2012	2012	2004
Reactor temperatures, °F.														
Maximum	1415	1405	1405	1410	1410	1510	1500	1505	1405	1390	1435	1425	1410	1415
Average	1240	1315	1350	1375	1385	1445	1465	1465	1265	1260	1260	1340	1305	1325
Char rate, lb./hr.	2.07	5.95	1.92	3.59	6.26	1.57	3.68	6.08	1.72	2.95	3.21	2.02	4.40	5.75
Hydrogen rate, SCF/hr.	34.48	34.87	66.50	65.54	66.37	65.50	68.02	69.81	93.15	92.57	102.84	130.26	132.90	132.90
Hydrogen/char ratio, SCF/lb.	16.68	16.86	34.65	18.25	10.60	41.85	18.47	11.19	52.88	24.10	31.43	64.69	29.59	23.12
Char space velocity, lb./cu.ft.-hr.	28.0	80.8	26.0	48.6	84.7	21.2	49.9	82.2	23.8	35.1	27.5	59.8	77.8	77.8
Hydrogen residence time, min.	1.35	1.34	1.33	1.38	1.34	1.29	1.23	1.18	1.49	1.47	1.31	1.34	1.36	1.55
Operating results														
Product gas rate, SCF (dry)/hr.	30.38	37.97	52.82	51.61	54.79	49.50	54.50	51.57	81.28	76.34	77.57	100.36	89.63	92.41
Product gas yield, SCF (dry)/lb.	14.70	6.38	27.52	14.38	8.75	31.63	14.80	8.49	46.14	19.45	14.73	49.39	20.36	16.00
Product gas net B.t.u. recovery, M B.t.u./lb	2.99	1.79	4.53	3.16	2.11	5.19	3.63	2.20	5.46	3.65	3.37	5.62	4.08	3.58
Gaseous hydrocarbon space-time yield, SCF (dry) cu.ft.-hr.	148	199	252	293	335	267	359	380	261	394	505	407	601	669
Net moisture-, ash-free char	26.4	16.3	42.7	29.3	21.1	52.9	32.4	21.3	48.2	32.5	30.3	58.0	40.6	35.5
Carbon gasified, wt. %	23.7	12.1	42.7	28.4	17.7	53.5	30.6	20.9	47.2	32.1	30.5	63.9	43.4	38.2
Feed hydrogen reacted, SCF (dry)/lb.	7.96	2.62	17.62	10.51	6.28	23.89	11.66	8.16	18.57	12.61	12.34	30.74	19.91	16.26
Char residue, lb./lb.	0.687	0.706	0.483	0.650	0.727	0.414	0.699	0.700	0.455	0.609	0.612	0.260	0.501	0.605
Condensed liquid products, lb. B.t.u. moisture, lb./lb.	0.0170	0.0031	0.9551	0.0201	0.0174	0.0681	0.0294	0.0395	0.0349	0.0759	0.0381	0.0685	0.0578	0.0445
Residue moisture, lb./lb.	0.0364	0.0264	0.0207	0.0531	0.0500	0.0259	0.0511	0.0364	0.0397	0.0410	0.0378	0.0282	0.0201	0.0326
Material balance, %	96.6	88.8	97.0	98.5	98.8	100.0	107.4	97.5	99.6	102.4	97.5	93.7	96.4	101.1
Product gas properties														
Gas composition, mole %														
N <sub>2</sub>	2.7	4.0	1.5	2.4	2.5	1.6	2.9	0.7	1.2	2.1	1.7	0.7	1.3	2.5
CO	1.3	2.9	1.5	1.5	2.5	1.5	2.1	5.2	2.0	0.6	2.0	0.4	1.4	1.2
CO <sub>2</sub>	59.2	52.3	0.1	0.3	0.4	0.2	0.2	0.5	0.1	0.2	0.1	0.1	0.2	0.2
H <sub>2</sub>	27.4	38.4	61.9	53.9	49.4	56.8	46.1	39.2	74.4	59.0	48.1	68.8	47.6	42.6
CH <sub>4</sub>	20.4	38.4	34.5	39.1	44.7	39.9	48.7	54.2	23.1	37.5	47.7	29.2	48.8	53.1
C <sub>2</sub> H <sub>6</sub>	0.2	0.2	0.4	1.4	0.5	--	--	0.1	0.5	0.2	0.2	0.6	0.6	0.3
Benzene	0.2	0.2	0.2	0.2	0.2	--	--	0.1	0.1	0.2	0.2	0.2	0.2	0.1
Total	100.6	100.6	100.2	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0
Heating value, B.t.u./SCF (ast.)	562	568	560	561	562	564	559	567	482	580	646	530	661	678
Specific gravity, air = 1	0.295	0.370	0.275	0.352	0.374	0.294	0.354	0.396	0.207	0.269	0.343	0.234	0.343	0.369

- a. Operating conditions and results based on weight of dry char
- b. cu.ft./min. hydrogen at reactor pressure and temperature
- c. 100 wt. of product gas - wt. of hydrogen in wt. of moisture-, ash-free char
- d. Hydrocarbons + water

REACTOR TEMP = 1400°F BED HEIGHT = 4.7 FT. BED VOL. = 0.074 CU. FT.  
 PRESS., P.S.I.G. = 500  
 H<sub>2</sub> RATE, SCF/HR. =  $\circ$  { 34-35 }  $\bullet$  { 64-67 }  $\Delta$  { 61-67 }  $\square$  { 93-102 }  $\nabla$  { 130-133 }  
 H<sub>2</sub> RES. TIME, MIN. = { 1.3-1.4 } { 0.7 } { 1.3-1.5 } { 1.3-1.5 } { 1.3-1.4 }

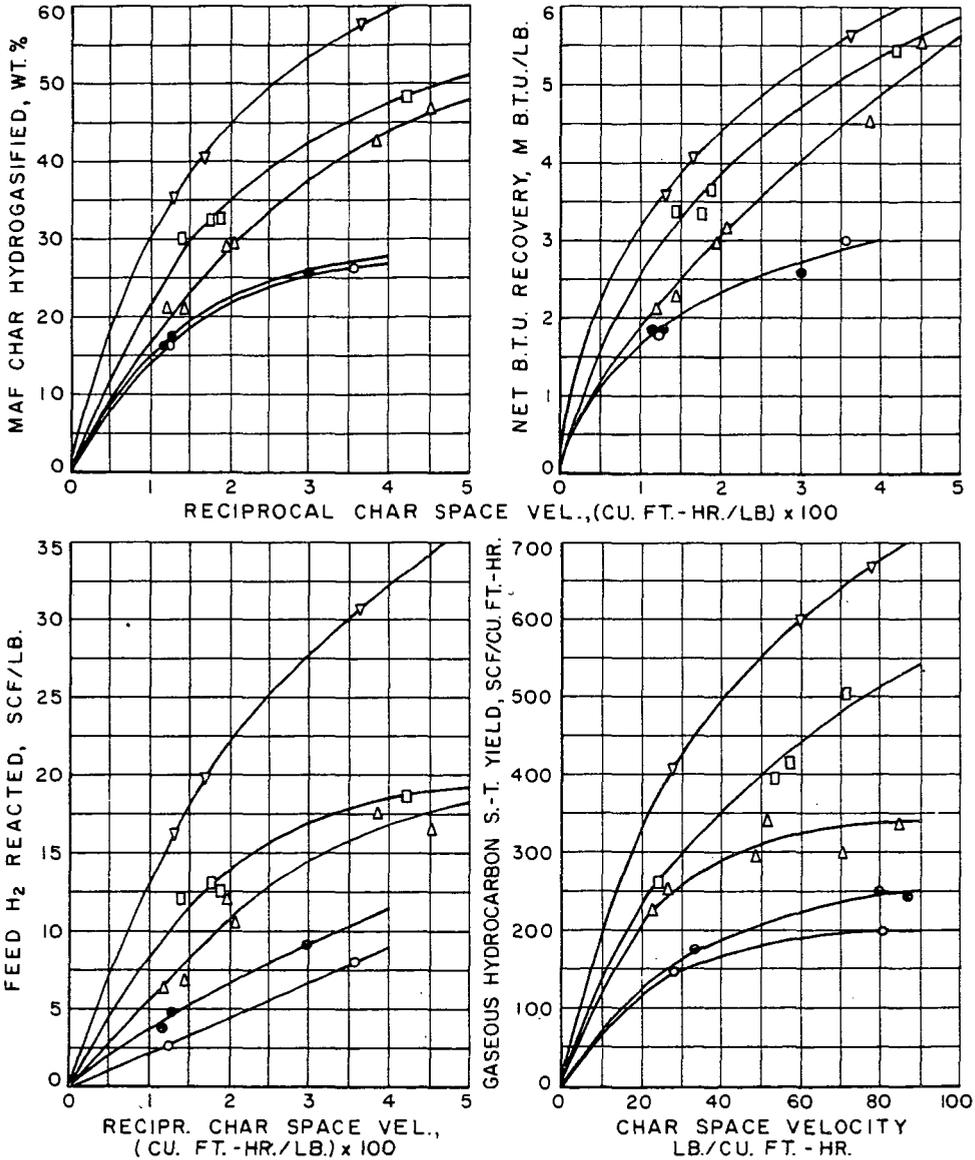


Fig. 3.-FLUID-BED HYDROGASIFICATION RESULTS WITH LOW-TEMPERATURE CHAR

(primarily methane) space-time yield with increases in pressure and increases in char space velocity. The increases in char conversion are accompanied by equivalent increases in the quantity of feed hydrogen reacted - the difference between the amount of hydrogen fed and the amount of unreacted hydrogen contained in the product gas, both per pound of dry char. The net B.t.u. recovery - the difference between the heat of combustion of the product gas and the heat of combustion of the feed hydrogen, both per pound of dry char - also exhibits the same trends as the conversion of char to gas. The data at 500 p.s.i.g. further show that an increase in hydrogen rate to double the level required to maintain a minimum superficial velocity results in a measurable increase in char conversion, gaseous hydrocarbon space-time yield, and quantity of feed hydrogen reacted. Similar results were obtained at higher pressures when the hydrogen feed rate was increased.

The effect of an increase in temperature from 1400° to 1500°F. is indicated in Table 2 in a series of runs at 1000 p.s.i.g. at each of the three feed rate levels. Except for the somewhat erratic behavior of the net B.t.u. recovery, which is an exceedingly sensitive parameter since it is obtained from a relatively small difference of two experimentally determined quantities, this increase in temperature resulted in a small but significant improvement in hydrogasification results.

The char feed and residue analysis data of Table 1 show that, in the course of hydrogasification, the volatile matter and oxygen-plus-nitrogen contents of the char were reduced to a small fraction of the original values, hydrogen content was reduced to about one-half of the original value, and sulfur content remained about the same. The increase in moisture content was caused by water formation through evolution of bound water or reactions of oxygen-containing coal constituents and gasification products with hydrogen; most of this water condensed in the residue receiver and moistened the char residue. Systematic variations of residual char properties with hydrogasification conditions appeared to exist, such as more severe reduction in hydrogen and volatile matter contents at the higher reaction temperature. The screen analyses of the feed and residual chars indicate some size reduction; however, analyses of the residues remaining in the reactor after shutdown showed this was not due to accumulation of larger particles in the reactor.

In the product distributions reported in Table 2, the condensed liquid products consisted primarily of water. Material balances can be seen to be near 100%, and the two parameters of net moisture-, ash-free char hydrogasified and carbon hydrogasified follow similar trends and are of approximately the same magnitude. In conjunction with the consistent behavior of the operating results over the range of test conditions employed, this indicates reliability of the reported data, in spite of the uncertainties introduced by fluid-bed operation at extremely low superficial gas velocities.

#### DISCUSSION OF RESULTS

The data obtained with low-temperature bituminous coal char and a 5-foot bed height show that a more reactive feed material, a deeper bed, or a number of countercurrent stages will be necessary to achieve both high conversions and high product gas heating values in a single-pass fluid-bed operation. However, an alternate technique for the production of a high-methane-content gas, comprising a hydrogen separation and recycle step, may be more attractive in spite of the

additional process steps required; the gasification rate would be increased by the resulting increase in hydrogen partial pressure, and problems of exothermic heat removal and control of gas-solids contacting greatly simplified. After completion of the present fluid-bed study, moving-bed operation allowing truly countercurrent single-pass contacting may also have to be considered.

Of considerable practical significance are the unexpectedly high gaseous hydrocarbon space-time yields obtained with the bituminous char at 1400°F. A series of studies by Gorin and others<sup>10,11,16,17</sup> of the kinetics of Disco char gasification with H<sub>2</sub>-steam mixtures and pure H<sub>2</sub> in a fluid-bed batch reactor at pressures up to 30 atmospheres (426 p.s.i.g.) and temperatures of 1500°-1700°F. are of interest in this connection. Direct comparison is not possible, since results are reported as differential gasification rates, i.e., rates corresponding to zero bed weight, which is equivalent to conditions where no equilibrium hindrance or other inhibiting effects of reaction products exist. The integral gasification rates obtained in this study would, therefore, be expected to be lower than equivalent differential rates, although this difference should be relatively small at low conversions. It appears that, due to the lower reactivity of Disco char, differential rates of methane formation extrapolated to the conditions of the present study are substantially lower than the integral rates reported in Table 2. For example, in terms of the units employed by Gorin and others, integral rates of methane formation in the 5-foot fluid bed at 1500°F. and 1000 p.s.i.g. ranged from about 70-100 x 10<sup>-4</sup> lb. moles CH<sub>4</sub>/lb. atom C per minute over a range of about 50-20% carbon gasification; this is approximately equivalent to extrapolated differential rates for Disco char and pure hydrogen at 1600°F.

At the relatively low pressures and high temperatures employed in the Disco char gasification study, steam was found to greatly accelerate methane formation, in addition to increasing char gasification by reactions leading to the formation of CO, CO<sub>2</sub> and H<sub>2</sub>. However, the effect of steam on methane formation decreased rapidly with increases in pressure. This was confirmed in an exploratory fluid-bed test at 1000 p.s.i.g., 1500°F. and 1.1:1 H<sub>2</sub>/steam mole ratio, in which the gaseous hydrocarbon space-time yield was 12% lower than in a test at the same conditions with H<sub>2</sub> feed only; a small increase in the rate of gaseous hydrocarbon formation was indicated by comparison with extrapolated data at the same inlet H<sub>2</sub> partial pressure.

The large increase in gaseous hydrocarbon space-time yield with increases in char feed rate, while the hydrogen rate was held nearly constant at each pressure level (Figure 3), occurred in spite of increased equilibrium hindrance from product methane. Since there was no clearly defined tendency for a shift in the source of the hydrogen contained in the gaseous hydrocarbons, this must have been caused primarily by an increase in average reactivity of the char bed. The quantity of feed hydrogen reacted increased nearly in proportion to the gaseous hydrocarbon formation, and corresponded to 80-100% of the hydrogen contained in the gaseous hydrocarbons over the whole range of operating conditions, except for some tests at 500 p.s.i.g. Conversely, the relatively small change in residual char properties with increases in char feed rate and pressure (Table 1) is consistent with the assumption that the increase in the amount of hydrogen supplied by the char was also roughly proportional to the increase in gaseous hydrocarbon formation.

#### CONCLUSIONS

The feasibility of producing high-methane-content fuel gas by continuous fluid-bed hydrogenolysis of a low-temperature bituminous coal char was demonstrated. In spite of the relatively low reactivity

of the feed material, it was possible to achieve about 40% conversion in a 5-foot bed at 2000 p.s.i.g. and 1400°F. when producing an approximately 50 mole % methane content gas. At these conditions, the gaseous hydrocarbon (mostly methane) production rate was about 600 SCF (dry)/hr.-cu.ft. bed volume, which is sufficiently high to be of commercial interest. Larger conversions were obtained at pressures as low as 1000 p.s.i.g., but at greatly reduced char space velocities, resulting in lower methane production. No major difficulties were encountered in operation of the fluid-bed reactor at an inlet hydrogen superficial velocity of only 0.06 ft./sec. at 500-2000 p.s.i.g. and 1400°-1500°F. Significant improvements in results are expected with deeper beds and more reactive feed materials.

#### ACKNOWLEDGMENT

This study was supported by the Gas Operations Research Committee of the American Gas Association with funds provided by the Promotion-Advertising-Research Plan of the Association. The guidance and counsel of the Project Supervising Committee under the chairmanship of B. J. Clarke, of T. L. Robey and N. K. Chaney of the American Gas Association, and of M. A. Elliott, director of the Institute of Gas Technology, were very helpful. S. Volchko was primarily responsible for the operation of the pilot unit. Analytical work was done under the supervision of D. M. Mason and J. E. Neuzil.

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