PILOT PLANT DEVELOPMENT OF THE HOT-GAS-RECYCLE PROCESS FOR THE SYNTHESIS OF HIGH-T.T.U. GAS. POROUS STAINLESS STEEL FILTERS FOR REMOVING DUST FROM HOT GASES

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PILOT PLANT DEVELOPMENT OF THE HOT-GAS-RECYCLE PROCESS FOR THE SYNTHESIS OF HIGH-B.T.U. GAS

By D. Bienstock, J. H. Field, A. J. Forney,
and R. J. Demski

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MARCH 3, 1849

UNITED STATES DEPARTMENT OF THE INTERIOR
Stewart L. Udall, Secretary

BUREAU OF MINES
Marling J. Ankeny, Director

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by

D. Bienstock,[2] J. H. Field,[2] A. J. Forney,[2] and R. J. Demski[3]

SUMMARY

A high-B.t.u. gas can be synthesized by the catalytic methanation of a mixture of 2.5 to 3 parts of hydrogen to 1 part of carbon monoxide, obtained in the gasification of coal, using a steel catalyst followed by Raney nickel in a hot-gas-recycle system. Conversion of 99 percent of the H_2+CO yields a fuel gas with a heating value of 983 to 1130 B.t.u. per cubic foot containing 78 to 96 percent methane, no carbon monoxide, and very small amounts of unsaturated hydrocarbons. With the steel catalyst alone, conversions were 85 to 90 percent, giving a fuel gas of 810 to 868 B.t.u. per cubic foot, or 847 to 975 B.t.u. on a CO_2-free basis.

INTRODUCTION

In 1948 the U.S. marketed production of natural gas was 5.15 trillion cubic feet; by 1958 it had reached 11.0 trillion cubic feet, a rate of growth of 8 percent annually.[4] [5] Although demand is expected to increase less rapidly, by 1966 it may reach 16 trillion cubic feet, based on an estimated 4.7 percent annual increase.[6]

Proved domestic reserves have increased from 173.9 to 252.2 trillion cubic feet from 1948 to 1958 because of new discoveries and extensions of existing fields, while the Life Index, the ratio of proved reserves to annual production, has dropped steadily from 29 to 21. This drop is influenced more by economic factors than by natural gas availability, and some of these factors are expected to make it profitable to produce a supplement for natural gas in some locations within the next decade.

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[3] Supervisory engineering technician, Bureau of Mines, Pittsburgh, Pa.

[4] Gas Age, The Supply of Natural Gas, A Review, Based on testimony of A. E. Kahn before the Federal Power Comm.: Vol. 122, No. 1, 1958, pp. 13-16.

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Synthesis of high-B.t.u. gas from coal offers a method of supplementing gas supplies. High-B.t.u. gas can be obtained either directly by hydrogenating coal or indirectly by gasifying coal to hydrogen and carbon monoxide followed by synthesis to gaseous hydrocarbons. This report describes a pilot plant operation by the Bureau of Mines at Bruceton, Pa., for the synthesis step in the catalytic hydrogenation of carbon monoxide. Part of this work was done in cooperation with the Consolidated Natural Gas Co., Pittsburgh, Pa.

Problems involved in the synthesis of gaseous hydrocarbons from carbon monoxide and hydrogen are similar to those for producing liquid hydrocarbons by the Fischer-Tropsch process. An active and durable catalyst is required, and the reactor design must provide for adequate removal of the exothermic heat of reaction. Heat liberated in synthesis of either gaseous or liquid hydrocarbons is about the same, approximately 65 B.t.u. per cubic foot of reactant when water is the oxidation product, and about 75 B.t.u. when carbon dioxide is formed. Whereas operating temperatures of 300° C. and lower are more favorable for producing liquid hydrocarbons, temperatures up to 400° C. have been employed for methanation.

For several years the Bureau of Mines has been studying the synthesis of liquid hydrocarbons using reactors of various designs. Liquid-phase processes, such as oil circulation[7] and slurry,[8] use a cooling oil in contact with the catalyst for heat removal. Well suited for producing liquid hydrocarbons, these processes are less applicable for producing methane; cracking and volatilization of the cooling oil occur at the high operating temperatures required. Dry-phase processes that are suitable for synthesis at high temperatures are the fixed-bed, fluidized and entrained processes. Both fixed-bed and fluidized reactors have been used in methanation studies by the Bureau,[9] [10] the Institute of Gas Technology,[11] and the British Gas Research Board.[12] An entrained fluidized system for synthesis of liquid hydrocarbons and oxygenates has been in commercial operation for several years.[13]

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[11] Dirksen, H. A., and Linden, H. R., Pipeline Gas by Methanation of Synthesis Gas over Raney Nickel Catalyst: Ind. Eng. Chem., vol. 52, 1960, pp. 584-589.

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A hot-gas-recycle process using a fixed bed of catalyst for the Fischer-Tropsch reaction has been under development at the Bureau of Mines for 3 years. In this process the heat of the exothermic reaction is removed by circulating large volumes of recycle gas through the catalyst bed. The sensible heat of the circulating gas is then recovered externally in a waste-heat boiler. This type of operation was applied by W. Michael[14] to the Fischer-Tropsch synthesis in 1938. To limit the rise in temperature of the catalyst bed to a maximum of 10° C., a ratio of recycle to fresh gas of 100 to 1 was employed. An economic deterrent to the hot-gas recycle as developed in Germany was the resistance to gas flow of the catalyst bed, a condition aggravated by the cubic shape of the catalyst employed. The power expended in circulating the large volumes of gas was prohibitive unless the catalyst bed was limited to rather shallow depths of 2 to 3 feet. Because the diameter of the reactor was restricted by pressure considerations, a large number of parallel reactors consuming an excessive amount of steel would have been necessary.

Steel in the form of lathe turnings has been successfully used as a catalyst in the oil-circulation process.[15] These turnings not only possess superior physical and chemical stability, but because they pack with a large void volume, 83 to 95 percent as compared with 50 percent for granular catalysts, they offer a lower resistance to gas flow. Smaller recycle flows can be used because of the good heat transfer characteristics of the turnings which, with their lower resistance, result in a considerable reduction in pressure drop. Figure 1 compares pressure drops of a granular magnetite with those of lathe turnings.[16] The pressure drop per foot of bed height, measured at the same conditions of flow, of 6- to 20-mesh iron oxide with a 48-percent void volume is 15 times greater than that of a bed of lathe turnings of 88-percent voids and 22 times greater than that with 94-percent voids.

From 1956 to 1958 a pilot plant using hot-gas recycle with lathe-turning catalyst has been operated successfully at the Bureau of Mines for synthesizing liquid hydrocarbons. With this background of operating experience, use of the hot-gas-recycle system was selected for methanation studies.

The specifications tentatively set for the high-B.t.u. gas are as follows:

1. Heating value of 900 B.t.u. per cubic foot or higher.
2. Carbon monoxide of 0.1 percent or less.
3. Not more than a trace of unsaturated hydrocarbons.
4. Optionally, enough ethane, propane, and butane to bring the heating value to 1100 to 1200 B.t.u. per cubic foot.

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[14] Faragher, W., and Foucher, J., The Hydrocarbon Synthesis from $CO+H_2$ on Iron Catalysts: Vol. 1, part C, 1947 FIAT Final Rept. 1267, PB 97,368, 123 pp.

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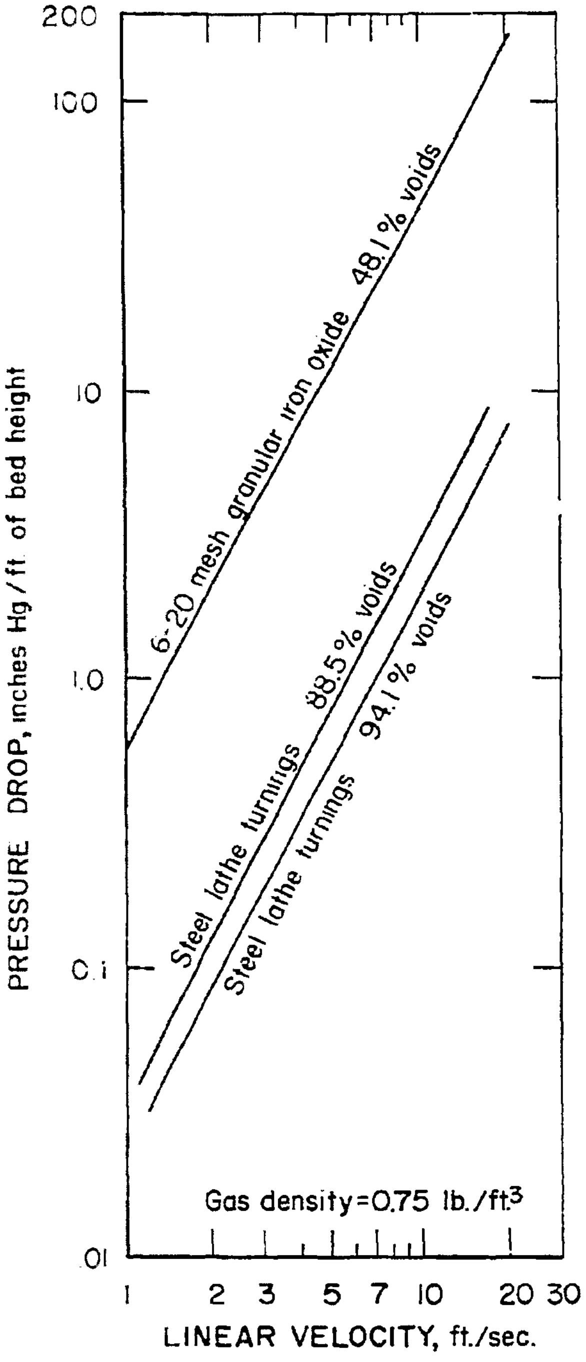

FIGURE 1. - Pressure Drops Through Beds of Various Catalyst Shapes.

EXPERIMENTAL PROCEDURE

Description of Apparatus

The major pieces of equipment include oxidation units for preparing the catalyst and a pilot plant for the synthesis.

Oxidation

A porous, catalytically active surface is prepared on the turnings by partial oxidation with steam followed by reduction with hydrogen.

The oxidizer for the steel turnings is a 6-inch Schedule 40 pipe, 2 feet in height fabricated from 304-stainless steel. Stainless-steel tubing, 3/8-inch outside-diameter, is coiled in a spiral around the oxidizer and serves as a steam preheater. The oxidizer and steam preheater are enclosed in a furnace which is electrically heated. (See fig. 2.) To obtain the higher temperatures required in oxidizing nickel turnings, a gas-fired unit with a 310-stainless steel reactor, 6 inches in diameter, was used. (See fig. 3.)

The effluent gases from the oxidizer pass through a water-cooled condenser and liquid-level device to condense and remove the steam. The non-condensible gases are dried, metered, and analyzed. The hydrogen production of the system is a measure of the degree of oxidation achieved.

Synthesis

There are two synthesis reactors. The bulk of the synthesis, 80 to 90 percent conversion of the feed gas, occurs in the main reactor over a steel catalyst; the remainder of the synthesis occurs in the second reactor over a Raney nickel catalyst.

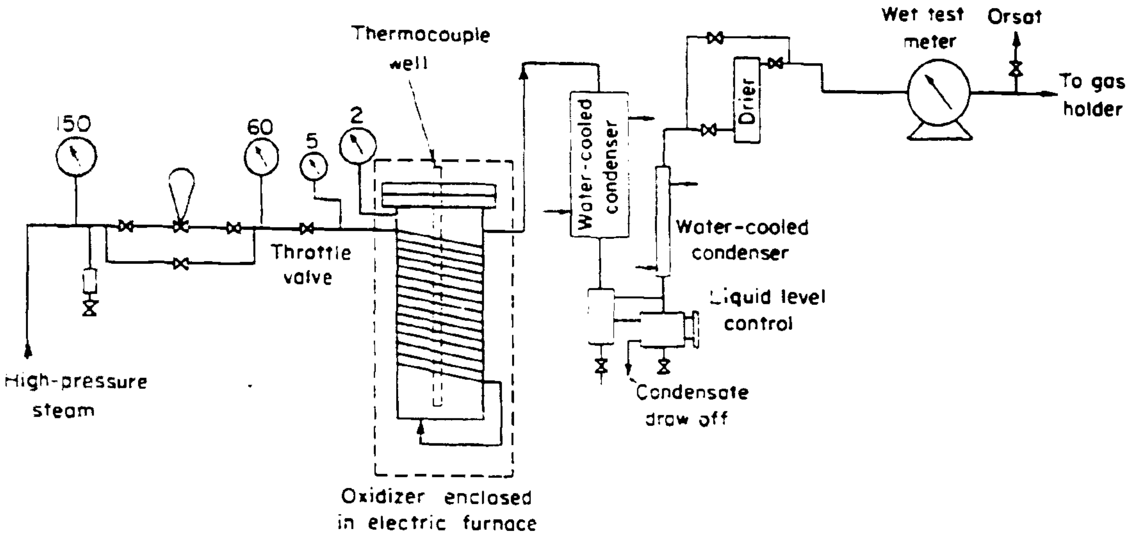

FIGURE 2. - Apparatus for Oxidation of Steel Turnings.

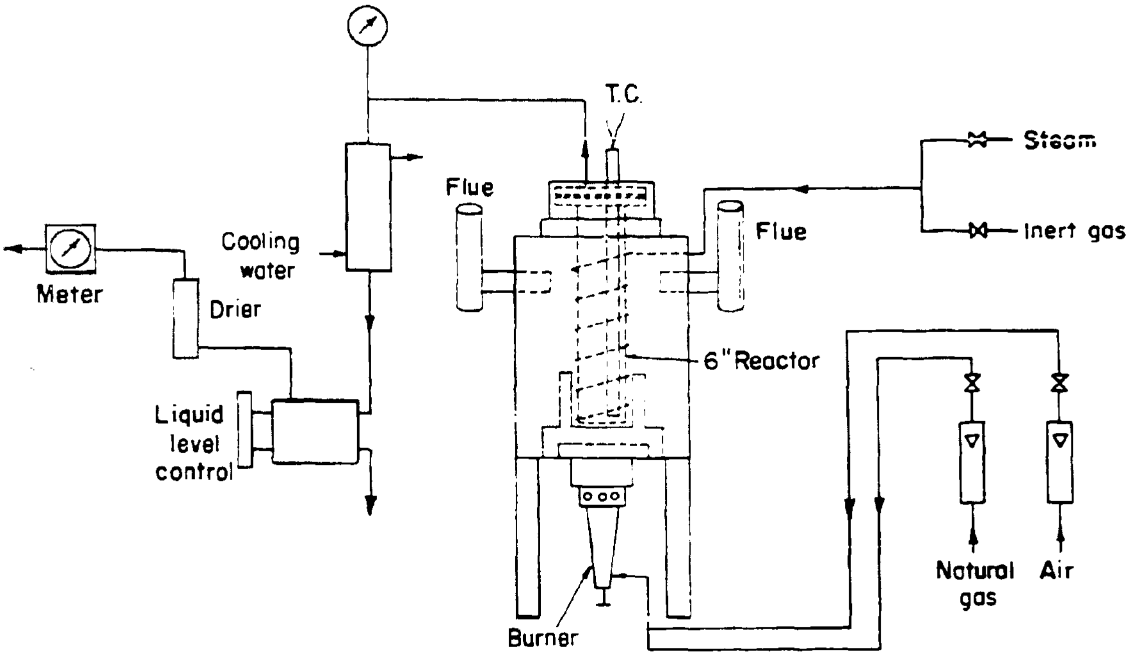

FIGURE 3. - Apparatus for Oxidation of Nickel Turnings.

Figure 4 shows the hot-gas-recycle system. Synthesis gas stored in a 60,000 cubic foot holder is compressed, dried, and then purified over activated carbon; its sulfur content is continuously analyzed and recorded. The fresh feed is metered, mixed with the cold recycle, and preheated indirectly by 150-pound steam. The combined stream is then mixed with the hot recycle.

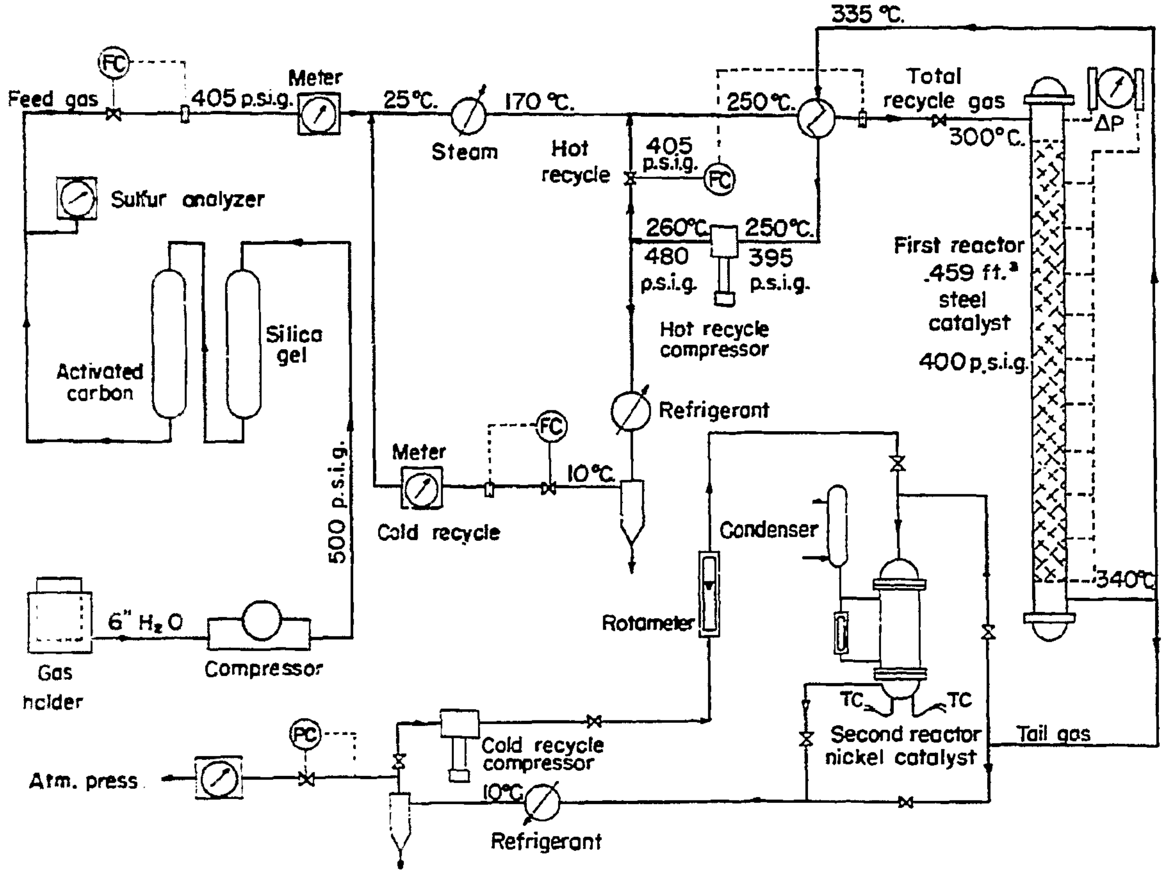

FIGURE 4. - Synthesis of High-B.t.u. Gas in the Hot-Gas-Recycle System.

The total feed, heated to reactor inlet temperature in the gas-to-gas exchangers, flows downward through the reactor. The gas leaving the reactor splits into tail gas and recycle gas streams. The recycle gas is cooled by heat exchange with the total feed and then recompressed. A small part of the recompressed gas, equivalent to 0.5 to 6 volumes of fresh feed, is cooled to 10° C. (the higher boiling hydrocarbons and water are condensed), metered, and returned as cold recycle. The major portion is returned as hot recycle. If one reactor is on stream, the tail gas is cooled and metered. If two reactors are in series, the tail gas is sent to the second reactor. All the exit gas of the second reactor is cooled, part is recompressed and returned as cold recycle to the second reactor; the remainder, the product gas stream, is metered and analyzed. When it is necessary to reduce the catalyst, hydrogen is substituted for synthesis gas. Before it enters the main reactor the hydrogen is heated to 400° C. by boiling Dowtherm.[17]

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[17] Reference to specific commercial products used in this research does not imply endorsement of the products by the Bureau.

Reactor

The main reactor is a 3-inch, Schedule 80, carbon-steel pipe, 12 feet long, provided with thermocouples and pressure taps at 1-foot intervals. The catalyst is supported on an 8-mesh stainless-steel screen located a few inches above the gas outlet. Beds as much as 10 feet high can be accommodated. A magnetic sampler can be lowered through the top of the reactor to withdraw samples of catalyst from the bed during synthesis. Figure 5 shows a cross section of the main reactor.

The second reactor consists of a 5-inch pipe in which four 1-inch diameter tubes containing the Raney nickel are placed. The catalyst tubes are surrounded by boiling Dowtherm, and thermocouple wells as shown in figure 6 are inserted axially within the tubes. A recycle of cold gas was used to assist in the heat removal, a recycle-to-feed gas ratio of 4 to 5 being used. This resulted in a temperature spread of 30° C. across the bed of nickel granules.

Hot-Gas-Recycle Compressor

The compressor for recycling the hot gases is a vertical, single-stage, double-acting, vertical gas booster designed to handle 27,000 standard cubic feet per hour of gas from an inlet pressure of 400 p.s.i.g. to an outlet pressure of 500 p.s.i.g. at 300° C. For large-scale operation, booster compressors of large flow capacity are readily available. A sulfur-free lubricant, Ucon LB-1715, was used for the overall lubrication--packing, cylinder walls, and bearings. Figure 7 shows the compressor.

Sulfur Removal

The synthesis gas used for the feed was prepared by reforming natural gas. The sulfur content, which was quite low, was further reduced by passing the gas over activated carbon yielding a feed gas whose sulfur content was less than 0.01 grain per 100 cubic feet.

Preparation of Catalyst

Steel Turnings

A 1018-carbon steel bar was cut on the lathe to produce turnings containing 88 percent voids. The turnings were oxidized with steam at an hourly space velocity of 400, a temperature of 600° C., and atmospheric pressure until approximately 20 percent of the iron was oxidized to Fe_3O_4 (magnetite). A batch of 26.43 pounds of the oxidized turnings was charged to the synthesis reactor to a height of 10 feet, 0.46 cubic foot, and was completely reduced by recycling hydrogen through the bed at an hourly space velocity of 4,000, a temperature of 400° C., and 100 p.s.i.g. for 60 hours. The steel turnings were used as such in experiments 18, 20, and 21. In experiment 21 they were impregnated with alkali before being reduced.

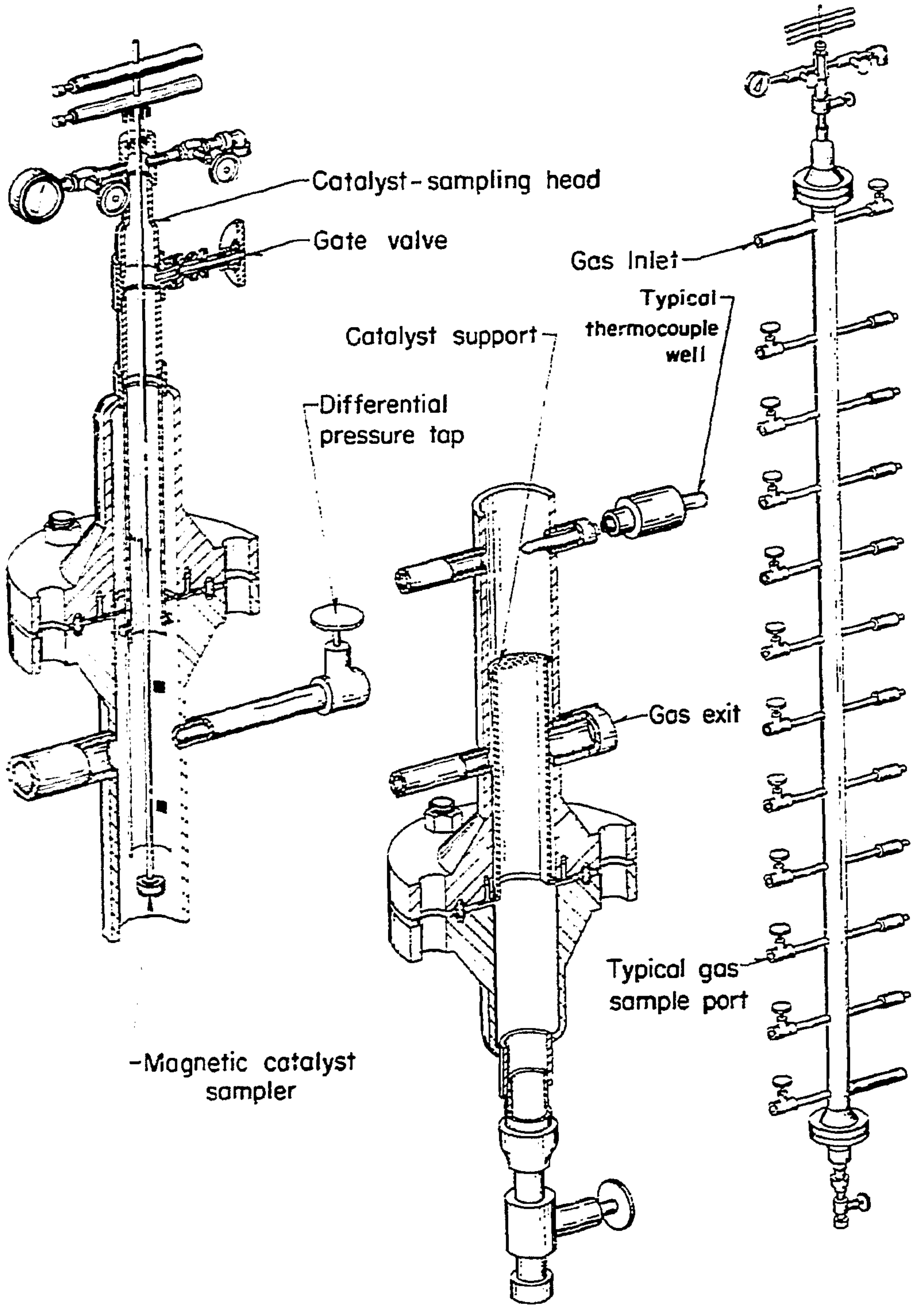

FIGURE 5. - Main Reactor, Hot-Gas-Recycle System.

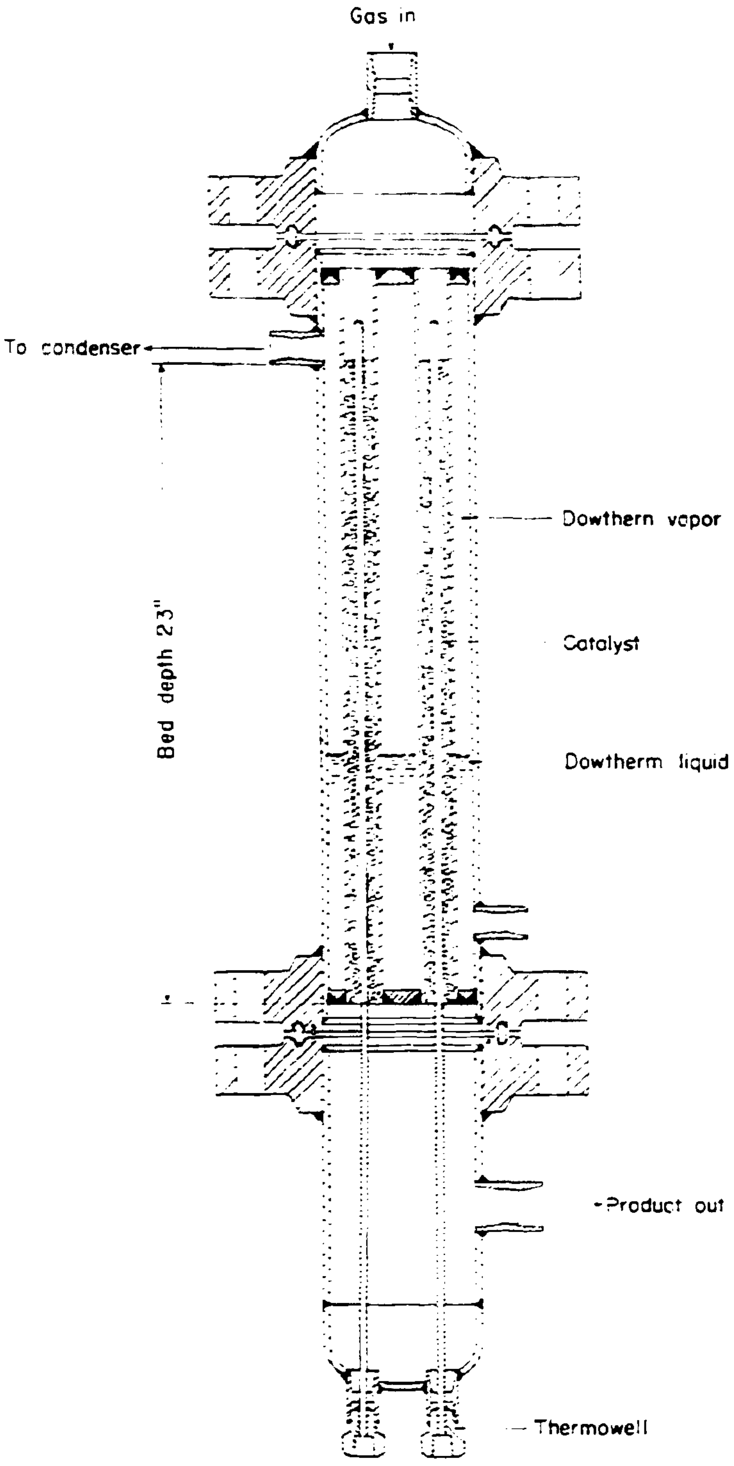

FIGURE 6. - Reactor 2, Hot-Gas-Recycle System.

Raney Nickel

The Raney nickel was activated by leaching it with a solution of sodium hydroxide. Raney nickel granules, 4- to 10-mesh, nominally 58 percent aluminum and 42 nickel, were added to a 25-percent solution of sodium hydroxide. Because considerable heat is released by the exothermic reaction, the granules were added at a controlled rate to prevent boiling the solution. Digestion was continued for an additional 2 hours. The alkali was decanted, and the partly extracted granules were washed with water. The wet granules, 4- to 20-mesh, were charged to the second reactor under an inert atmosphere and dried with hydrogen at 300° C. for 24 hours before use.

Nickel Turnings

In experiment 19, nickel turnings were used instead of the steel turnings. Turnings were cut from a commercial nickel bar and packed with a void volume of 93 percent. They were oxidized with steam at 900° to 950° C. until 18.3 percent of the nickel was converted to the oxide. A batch of 18.75 pounds of the oxidized nickel turnings was charged to the main reactor to a height of 10 feet and reduced by recycling hydrogen through

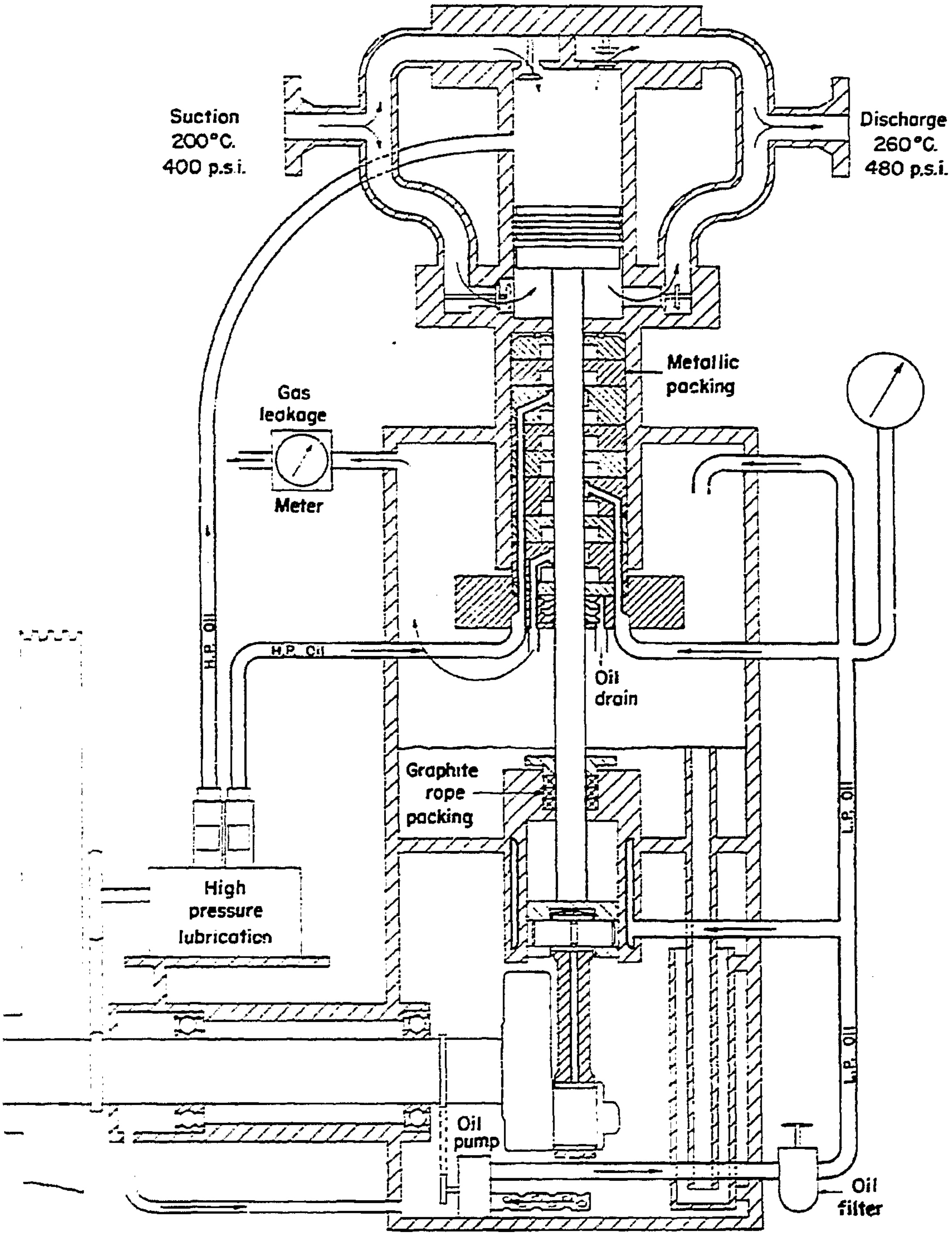

FIGURE 7. - Recycle Compressor, Hot-Gas-Recycle System.

the bed at an hourly space velocity of 4000, a temperature of 400° C., and 100 p.s.i.g. for 60 hours.

Synthesis of High-B.t.u. Gas

Synthesis was started over the reduced steel turnings in the first reactor as follows: A reactor pressure of 400 p.s.i.g., a fresh feed hourly space velocity of 200, and a recycle-to-fresh feed ratio of 40 to 1. The reactor temperatures were increased in steps: To 200° C. in 4 hours, from 200° to 265° C. in 15 hours, and from 265° to 320° C. in 24 hours. The hourly space velocity of fresh feed was gradually raised to 800 to 1,000.

Operating conditions were arranged so the following factors could be investigated: (1) Fresh-feed ratios of H_2 to CO of 2.5, 2.7, and 3.0; (2) cold recycle to fresh feed ratios of 6, 4, 2, 1, 0.5, and 0; (3) operations with one and both reactors; (4) alkali impregnation of the steel turnings, and (5) use of nickel turnings.

Operating conditions and results of these tests are listed in tables 1, 2, and 3. Experiments 18, 20, and 21 were voluntarily terminated at the end of 640, 1,204, and 2,314 hours of operation, respectively, with the catalyst retaining much of its original activity.

DISCUSSION OF RESULTS

Single Reactor with Steel-Turning Catalyst

Total Gas Recycle

With a fresh feed at an hourly space velocity of 800 to 1,000 and conversions of 85 to 90 percent, excellent temperature control of the bed could be maintained when the total recycle-to-fresh gas ratio was as low as 20. At these conditions a temperature differential of 40° C. was obtained across the bed, with an average temperature of 320° C. No hot spots occurred at any time. No attempt was made to operate the equipment at lower recycle ratios.

Synthesis-Gas Composition

Fresh feed with hydrogen-to-carbon monoxide ratios of 2.5, 2.7, and 3.0 was used at an hourly space velocity of 800 and a cold recycle-to-fresh gas ratio of 4. Figure 8 and table 4 show that the heating values of the product gas on a dry basis, without use of the second reactor, ranged from 810 to 868 B.t.u. per cubic foot. The maximum value was obtained with 2.5 gas ratio and the minimum with the 3.0 gas ratio. The higher heating value occurred despite the lower methane and higher carbon dioxide content because of the increase in the content of C_2 plus paraffin hydrocarbons. The olefins were constant at 0.6 percent. On a carbon dioxide-free basis the heating value ranged from 847 to 975. About 3.5 to 3.7 volumes of feed gas are required to produce one volume of product gas.

TABLE 1. - Experiment 18, hot-gas-recycle system

Period	2.7:1 Synthesis gas							
	A	B	C	D	E	F	G	H
Hours of synthesis	60-84	110-132	144-156	168-192	204-228	240-288	292-316	324-396
Number of reactors	1	1	1	2	1	2	2	2
Main reactor:								
Space velocity, $hr.^{-1}$	1000	1002	997	1000	802	801	798	801
Recycle:fresh feed	19	19	19	19	19	19	19	20
Cold recycle:fresh feed	2.2	4.0	6.1	6.0	4.0	4.0	6.0	2.0
Pressure, p.s.i.g.	398	400	400	400	400	400	398	400
Temperature, ° C.:								
In gas	300	300	300	300	300	300	300	300
Out gas	340	340	340	340	340	340	340	340
ΔT	40	40	40	40	40	40	40	40
Average	320	323	323	320	317	318	318	317
Pressure drop, inches H_2O per foot	13.3	12.9	12.5	16.1	19.4	27.2	30.2	31.2
Second reactor, average temperature, ° C.	-	-	-	365	-	395	400	400
Carbon dioxide-free contraction, percent	71.0	71.5	72.0	77.3	74.1	77.1	76.7	76.3
Hydrogen conversion, do.	79.2	81.9	83.1	99.6	86.6	99.3	99.3	98.7
Carbon monoxide conversion, do.	97.6	96.8	96.4	100	98.1	99.9	100	99.9
Hydrogen + carbon monoxide conversion, do.	84.2	85.9	86.7	99.7	89.7	99.4	99.5	99.0
Hydrogen:carbon monoxide ratio:								
Fresh feed	2.69	2.70	2.74	2.72	2.71	2.70	2.68	2.67
Recycle, main reactor	23.5	15.1	12.7	12.7	19.3	26.8	13.8	18.6
Usage	2.18	2.28	2.36	2.71	2.39	2.68	2.65	2.64
Water vapor, recycle main reactor, volume percent	7.4	4.4	3.3	3.2	4.9	4.8	3.5	8.3
Product gas, volume-percent (dry basis):								
H_2	47.0	42.4	40.6	1.2	34.7	2.2	2.0	3.7
CO	2.0	2.8	3.2	0	1.8	0.1	0	0.1
N_2	0.9	0.8	1.3	1.2	1.0	1.1	1.5	1.6
CO_2	9.5	8.5	7.4	6.9	8.1	7.3	7.4	7.7
C_1	29.1	32.6	36.1	89.9	40.5	88.4	87.9	85.8
$C_2=$	0	0	0	0	0	0	0	0
C_2	5.9	7.0	6.2	0.2	7.6	0.3	0.4	0.3
$C_3=$	0.3	0.3	0.3	0.2	0.3	0.1	0.2	0.1
C_3	3.4	3.8	3.2	0.3	4.0	0.3	0.4	0.3
$C_4=$	0.2	0.2	0.2	0	0.2	0.1	0.1	0.1
C_4	1.3	1.2	1.2	0.1	1.3	0.1	0.1	0.2
$C_5=$	0	0	0	0	0.1	0	0	0
C_5	0.4	0.4	0.3	0	0.4	0	0	0.1
$C_6=$	0	0	0	0	0	0	0	0
H_2O	7.4	4.4	3.3	11.4	4.9	10.5	7.6	14.2
Heating value product gas, $B.t.u./ft.^3$ [1]	721	771	768	934	846	925	926	911
Volume, fresh feed:volume, product gas	3.12	3.21	3.31	4.09	3.55	4.04	3.97	3.89
Yield, $g./m.^3$ H_2+CO conversion:								
C_1	79.1	84.5	91.2	156.9	91.2	157.4	158.8	159.7
$C_2=$	0	0	0	0	0	0	0	0
C_2	30.7	34.7	29.6	0.7	32.4	1.0	1.3	1.1
$C_3=$	2.2	2.1	2.0	0.9	1.8	0.5	1.0	0.5
C_3	26.3	28.0	22.8	1.5	25.4	1.5	2.1	1.6
$C_4=$	2.0	1.7	1.8	0	1.6	0.6	0.6	0.7
C_4	13.5	11.9	11.4	0.6	11.0	0.7	0.7	1.4
$C_5=$	0	0	0	0	1.0	0	0	0
C_5	5.2	4.9	3.6	0	4.2	0	0	0.8
$C_6=$	0	0	0	0	0	0	0	0
Oil	14.3	13.7	9.6	8.7	10.4	9.1	7.7	8.6
Aqueous phase	192.1	191.5	204.1	185.0	198.1	187.3	183.0	181.3
C_1-C_3OH [2]	1.3	1.9	2.8	2.8	1.6	1.6	1.8	0.9
Other oxygenates [3]	0.3	0.1	0.3	0.5	0.3	0.3	0.1	0.3
H_2O	190.5	189.5	201.0	181.7	196.2	185.4	181.1	180.1
CO_2	72.0	61.4	51.6	31.4	50.4	36.0	37.1	39.8
Hydrocarbon recovery, $g./m.^3$ H_2+CO conversion	175.1	184.0	175.0	172.7	181.5	173.0	174.5	175.9
Theoretical recovery, $g./m.^3$ H_2+CO conversion	190.2	189.2	188.4	182.2	188.2	182.1	181.6	181.8

See footnotes at end of table.

TABLE 1. - Experiment 18, hot-gas-recycle system (Con.)

Period	3:1 Synthesis gas				2.5:1 Synthesis gas			
	I	J	K	L	M	N	O	P
Hours of synthesis	420-468	476-492	496-508	516-540	552-576	580-604	612-624	628-640
Number of reactors	2	1	1	2	2	2	1	1
Main reactor:								
Space velocity, hr.$^{-1}$	802	802	803	797	798	800	800	805
Recycle:fresh feed	19	20	19	18	18	18	18	17
Cold recycle:fresh feed	2.0	2.0	4.0	4.0	4.0	2.0	2.0	4.0
Pressure, p.s.i.g.	400	404	406	400	400	401	401	399
Temperature, ° C.:								
In gas	300	301	300	300	300	300	300	300
Out gas	340	340	340	340	340	340	340	340
ΔT	40	39	40	40	40	40	40	40
Average	319	319	319	319	320	319	319	321
Pressure drop, inches H_2O per foot	29.4	29.4	29.5	28.9	33.3	33.9	33.2	33.1
Second reactor, average temperature, ° C.	390	-	-	400	395	385	-	-
Carbon dioxide-free contraction, percent	75.4	70.0	72.3	75.6	77.4	77.1	74.1	76.2
Hydrogen conversion, do.	98.5	80.6	84.9	98.4	99.1	99.2	85.3	88.2
Carbon monoxide conversion, do.	100	98.7	98.5	99.8	99.3	100	98.1	98.0
Hydrogen + carbon monoxide conversion, do.	98.9	85.3	88.4	98.7	99.2	99.4	89.0	91.0
Hydrogen:carbon monoxide ratio:								
Fresh feed	3.05	2.93	2.95	3.00	2.47	2.49	2.46	2.46
Recycle, main reactor	58.3	44.8	29.8	29.8	17.3	18.5	18.5	17.3
Usage	3.00	2.39	2.53	2.96	2.47	2.47	2.14	2.21
Water vapor, recycle main reactor, volume-percent	8.1	8.0	4.8	4.7	4.8	8.0	7.9	4.7
Product gas, volume-percent (dry basis):								
H_2	4.5	44.8	38.8	4.9	2.4	2.2	35.2	31.2
CO	0	1.0	1.3	0.2	0.8	0	1.9	2.2
N_2	1.4	1.0	1.3	1.2	0.7	1.5	1.5	1.1
CO_2	2.1	6.4	4.4	1.9	12.5	12.8	12.3	11.0
C_1	91.4	37.1	43.6	90.9	81.7	82.0	37.5	38.8
$C_2=$	0	0	0	0	0	0	0	0
C_2	0.2	5.4	6.0	0.5	0.6	0.5	5.9	7.0
$C_3=$	0.1	0.4	0.3	0.1	0.3	0.1	0.4	0.3
C_3	0.2	2.7	2.9	0.3	0.6	0.4	3.4	6.2
$C_4=$	0	0.2	0.2	0	0.1	0.1	0.3	0.3
C_4	0.1	0.9	0.9	0	0.3	0.3	1.3	1.7
$C_5=$	0	0	0.1	0	0	0	0.2	0.1
C_5	0	0.1	0.2	0	0	0.1	0.1	0.1
$C_6=$	0	0	0	0	0	0	0	0
H_2O	19.8	8.0	4.8	13.5	6.7	13.8	7.9	4.7
Heating value product gas, B.t.u./ft.^{3} [1]	955	741	810	956	884	877	768	868
Volume, fresh feed:volume, product gas	3.98	3.11	3.46	4.01	3.87	3.81	3.39	3.74
Yield, g./m.^{3} H_2+CO conversion:								
C_1	167.3	100.6	102.8	164.7	152.1	154.9	88.3	80.9
$C_2=$	0	0	0	0	0	0	0	0
C_2	0.7	27.7	26.7	1.7	2.1	1.8	26.5	27.9
$C_3=$	0.5	2.9	1.9	0.5	1.5	0.5	2.5	1.6
C_3	1.0	20.6	19.3	1.6	3.2	2.1	22.7	36.8
$C_4=$	0	2.0	1.7	0	0.6	0.7	2.5	2.2
C_4	0.7	9.2	8.0	0	2.1	2.1	11.7	13.5
$C_5=$	0	0	1.1	0	0	0	2.2	1.0
C_5	0	1.3	2.2	0	0	0.9	1.1	1.0
$C_6=$	0	0	0	0	0	0	0	0
Oil	4.5	5.9	5.5	5.3	13.7	13.2	15.3	16.6
Aqueous phase	187.2	193.1	197.4	195.6	177.0	174.5	173.0	189.1
C_1-C_3OH[2]	0.9	1.1	1.2	1.3	1.4	1.3	1.4	1.8
Other oxygenates[2]	0.1	0.1	0.1	0.3	0.3	0.4	0.4	0.4
H_2O	186.2	191.9	196.1	194.0	175.3	172.8	171.2	186.9
CO_2	10.6	47.9	28.6	9.5	64.6	67.1	80.6	63.9
Hydrocarbon recovery, g./m.^{3} H_2+CO conversion	176.1	171.7	170.8	175.6	177.2	178.3	174.8	183.9
Theoretical recovery, g./m.^{3} H_2+CO conversion	180.6	187.6	187.6	181.0	182.8	182.6	189.5	189.5

[1] Cubic foot corrected to 60° F. and 30 in. Hg, dry.

[2] Calculated as hydrocarbons.

TABLE 2. - Experiment 20, hot-gas-recycle system

Period	3:1 Feed								
	A	B	C	D	E	F	G	H	I
Hours of synthesis	93-129	153-201	225-273	301-369	472-520	532-568	580-616	648-688	692-716
Number of reactors	1	1	1	2	1	2	2	2	1
Main reactor:									
Space velocity ... hr.$^{-1}$	802	602	599	602	601	602	601	599	601
Recycle:fresh feed	19	19	19	19	19	19	18	19	19
Cold recycle:fresh feed	2.0	2.0	1.5	1.5	2.0	2.0	2.0	2.0	2.0
Pressure ... p.s.i.g.	402	402	402	402	401	401	400	402	402
Temperature, ° C.:									
In gas	300	300	300	299	300	299	300	300	300
Out gas	340	340	339	340	340	340	340	340	340
ΔT	40	40	39	41	40	41	40	40	40
Average	322	323	322	320	320	319	320	321	320
Pressure drop ... inches H_2O per foot	8.2	4.6	4.8	4.7	5.0	5.4	6.2	6.9	7.7
Second reactor, average temperature ... ° C.	-	-	-	300	-	300	270	350	-
Carbon dioxide-free contraction ... percent	63.8	64.5	62.6	76.6	56.7	77.0	76.4	76.0	61.7
Hydrogen conversion ... do.	70.0	71.5	68.6	99.0	63.6	98.9	93.5	99.8	70.0
Carbon monoxide conversion ... do.	95.7	97.7	94.2	100	90.0	100	99.9	100	94.3
Hydrogen + carbon monoxide conversion ... do.	76.6	78.0	75.1	99.2	70.2	99.0	95.2	99.9	76.1
Hydrogen:carbon monoxide ratio:									
Fresh feed	2.91	3.02	2.95	2.95	3.00	2.98	2.91	3.02	2.96
Recycle, main reactor	20.0	36.7	15.8	-	11.0	-	-	-	15.6
Usage	2.13	2.21	2.14	2.92	2.12	2.93	2.73	3.01	2.20
Water vapor, recycle main reactor, volume percent	7.0	7.2	8.5	8.5	5.6	6.0	6.1	6.4	6.9
Product gas, volume-percent (dry basis):									
H_2	56.2	55.1	56.9	3.2	57.0	4.5	20.0	0.6	53.1
CO	2.8	1.5	3.6	0	5.2	0	0.1	0	3.4
N_2	1.0	1.5	0.3	1.4	0.8	1.6	1.7	1.2	0.3
CO_2	8.2	7.9	8.8	1.0	8.9	0.6	1.1	1.8	9.0
C_1	23.1	26.0	22.8	92.7	23.1	89.5	67.1	56.0	27.5
C_2=	0	0	0	0	0	0	0	0	0
C_2	4.5	4.4	3.8	1.3	3.0	3.0	6.1	0.4	3.7
C_3=	0.3	0.2	0.3	0.1	0.3	0.1	0.3	0	0.3
C_3	2.5	2.2	2.2	0.3	1.3	0.6	2.7	0	1.8
C_4=	0.2	0.2	0.2	0	0.1	0	0.1	0	0.1
C_4	0.9	0.8	0.8	0	0.3	0.1	0.8	0	0.6
C_5=	0	0	0.1	0	0	0	0	0	0
C_5	0.3	0.2	0.2	0	0	0	0	0	0.2
C_6=	0	0	0	0	0	0	0	0	0
H_2O	7.0	7.2	8.5	30.6	5.6	32.9	32.0	30.0	6.9
Heating value product gas ... B.t.u./ft.^{3} [1]	627	629	605	983	544	996	961	983	613
Volume, fresh feed:volume, product gas	2.54	2.60	2.44	4.23	2.10	4.33	4.19	4.09	2.38
Yield, g./m.^{3} H_2+CO conversion:									
C_1	83.7	91.8	88.9	158.6	110.9	150.0	120.2	168.3	108.4
C_2=	0	0	0	0	0	0	0	0	0
C_2	31.6	29.7	28.3	4.2	27.8	9.6	20.8	1.3	27.9
C_3=	3.0	1.9	3.2	0.5	3.9	0.4	1.5	0	3.2
C_3	26.1	22.1	24.4	1.5	17.9	2.9	13.7	0	20.1
C_4=	2.7	2.5	2.9	0	1.7	0	0.8	0	1.4
C_4	12.5	10.7	11.9	0	5.5	0.6	6.8	0	9.1
C_5=	0	0	1.8	0	0	0	0	0	0
C_5	5.2	3.3	3.8	0	0	0	0	0	3.7
C_6-	0	0	0	0	0	0	0	0	0
Oil	11.3	8.6	8.1	4.8	4.4	2.8	2.2	1.6	3.3
Aqueous phase	181.3	185.8	179.1	197.1	169.6	200.4	201.0	191.8	173.1
C_1-C_3OH [1]	2.3	2.4	2.2	1.4	5.5	2.8	2.5	2.0	3.0
Other oxygenates [2]	0.3	0.3	0.4	0.3	0.5	0.4	0.3	0.3	0.2
H_2O	178.7	183.1	176.5	195.4	163.6	197.2	198.2	189.5	169.9
CO_2	81.2	77.7	93.0	4.7	117.2	0.7	3.4	8.7	96.2
Hydrocarbon recovery ... g./m.^{3} H_2-CO conversion	178.7	173.3	175.9	171.3	178.6	170.2	168.8	173.5	180.3
Theoretical recovery ... g./m.^{3} H_2-CO conversion	189.3	188.8	189.1	181.9	186.2	182.4	178.4	180.4	186.3

See footnotes at end of table.

TABLE 2. - Experiment 20, hot-gas-recycle system (Con.)

Period	2.7:1 Feed							
	J	K	L	M	N	O	P	Q
Hours of synthesis	736-760	768-808	816-856	864-904	984-1,024	1,032-1,072	1,096-1,144	1,156-1,204
Number of reactors	1	2	2	2	2	2	1	1
Main reactor:								
Space velocity ... hr.$^{-1}$	597	599	601	601	602	601	600	600
Recycle:fresh feed	19	19	19	19	19	19	20	20
Cold recycle:fresh feed	2.0	2.0	2.0	2.0	2.0	2.0	3.6	3.9
Pressure ... p.s.i.g.	401	402	403	400	402	400	400	399
Temperature, ° C.:								
In gas	300	300	299	300	300	300	260	309
Out gas	339	340	340	340	340	340	300	350
ΔT	39	40	41	40	40	40	40	41
Average	320	320	320	322	322	322	266	315
Pressure drop ... inches H_2O per foot	8.5	8.5	8.8	9.2	9.9	10.8	28.6	26.8
Second reactor, average temperature ... ° C.	-	270	300	350	350	300	-	-
Carbon dioxide-free contraction ... percent	65.2	78.2	80.1	80.0	71.4	70.1	73.4	71.5
Hydrogen conversion ... do.	73.7	94.5	98.4	99.7	87.9	80.5	92.3	93.4
Carbon monoxide conversion ... do.	94.5	99.7	100	99.9	99.7	98.6	95.0	96.8
Hydrogen + carbon monoxide conversion ... do.	79.3	95.9	98.8	99.7	91.0	85.3	93.0	94.3
Hydrogen:carbon monoxide ratio:								
Fresh feed	2.71	2.71	2.67	2.75	2.71	2.73	2.70	2.68
Recycle, main reactor	12.8	42.8	-	-	-	-	4.12	5.50
Usage	2.11	2.57	2.62	2.75	2.39	2.23	2.62	2.58
Water vapor, recycle main reactor, volume-percent	6.9	6.9	6.9	7.1	6.6	6.9	5.5	4.9
Product gas, volume-percent (dry basis):								
H_2	48.8	17.1	5.8	1.2	30.9	42.3	19.8	15.4
CO	3.8	0.4	0	0.1	0.3	1.2	4.8	2.8
N_2	0.9	1.3	1.3	1.8	1.2	1.2	0.2	0
CO_2	11.0	5.8	3.3	3.7	10.2	10.9	6.6	9.1
C_1	28.0	62.9	74.9	78.2	47.2	35.5	61.3	70.2
$C_2=$	0	0	0	0	0	0	0	0
C_2	4.1	7.2	8.5	8.6	5.6	4.8	4.7	1.8
$C_3=$	0.2	0.2	0.3	0.5	0.3	0.3	0.2	0.1
C_3	2.0	3.6	4.2	4.2	2.8	2.5	2.0	0.5
$C_4=$	0.2	0.1	0.1	0.1	0.1	0.1	0.1	0
C_4	0.7	1.3	1.3	1.3	1.1	1.0	0.3	0.1
$C_5=$	0.1	0	0	0	0.1	0	0	0
C_5	0.2	0.1	0.3	0.3	1.2	0.2	0	0
C_6+	0	0	0	0	0	0	0	0
H_2O	6.9	28.7	33.6	32.1	21.3	12.3	5.5	4.9
Heating value product gas ... B.t.u./ft.^{3} [1]	626	972	1,105	1,130	811	704	855	821
Volume, fresh feed:volume, product gas	2.56	4.32	4.87	4.82	3.50	2.98	3.51	3.18
Yield, g./m.^{3} H_2+CO conversion:								
C_1	99.0	108.9	111.6	116.4	105.9	99.9	133.7	167.4
$C_2=$	0	0	0	0	0	0	0	0
C_2	27.7	23.7	24.2	24.4	23.9	25.7	19.5	8.1
$C_3=$	1.9	0.9	1.2	2.0	1.8	2.3	1.2	0.7
C_3	20.1	17.7	17.8	17.7	17.8	19.9	12.4	3.3
$C_4=$	2.5	0.6	0.5	0.6	0.8	1.0	0.8	0
C_4	9.4	8.6	7.4	7.3	9.4	10.8	2.4	0.8
$C_5=$	1.6	0	0	0	1.1	0	0	0
C_5	3.2	0.8	2.1	2.1	2.2	2.6	0	0
C_6+	0	0	0	0	0	0	0	0
Oil	6.6	5.9	5.0	3.5	4.9	7.6	0	0
Aqueous phase	178.3	202.2	204.8	204.0	187.4	184.2	192.0	175.8
C_1-C_3OH [2]	2.8	1.8	1.8	1.8	2.1	2.1	1.4	1.1
Other oxygenates [2]	0.4	4.6	4.6	4.6	1.1	1.1	0.4	0.4
H_2O	175.1	195.8	198.4	197.6	184.2	181.0	190.2	174.3
CO_2	89.4	25.9	11.8	15.3	63.7	85.3	40.0	59.9
Hydrocarbon recovery ... g./m.^{3} H_2+CO conversion	175.2	173.5	176.3	180.4	171.0	173.0	171.8	181.8
Theoretical recovery ... g./m.^{3} H_2+CO conversion	187.9	186.8	186.5	185.3	187.2	188.0	183.9	180.3

[1] Cubic foot corrected to 60° F. and 30 in. Hg, dry.

[2] Calculated as hydrocarbons.

TABLE 3. - Experiment 21, hot-gas-recycle system

Period	A	B	C	D	E	F	G	H	I
Hours of synthesis	183-231	254-290	347-395	443-515	519-531	539-611	731-779	827-875	947-995
Number of reactors	1	1	1	2	1	1	2	2	2
Main reactor:									
Space velocity ... hr.$^{-1}$	1,002	1,001	999	1,001	1,000	1,002	1,003	999	1,005
Recycle:fresh feed	22	22	21	20	20	21	19	19	20
Cold recycle:fresh feed	2.0	2.0	2.0	2.0	2.0	1.0	1.0	1.0	1.0
Pressure ... p.s.i.g.	402	401	400	398	398	401	400	400	401
Temperature, ° C.:									
In gas	300	300	300	299	299	299	302	301	301
Out gas	341	340	341	340	340	339	342	341	341
ΔT	41	40	41	41	41	40	40	40	40
Average	320	320	319	319	319	318	320	318	316
Pressure drop ... inches H_2O per foot	11	13	9	9	9	10	11	11	10
Second reactor, average temperature ... ° C.	-	-	-	305	-	-	304	353	306
Carbon dioxide-free contraction ... percent	72.9	79.2	71.9	76.7	71.9	66.6	77.3	77.4	77.5
Hydrogen conversion ... do.	75.2	81.0	74.8	87.7	76.2	67.0	86.3	97.6	86.7
Carbon monoxide conversion ... do.	98.7	98.2	97.6	99.4	98.0	97.9	99.5	99.9	99.5
Hydrogen + carbon monoxide conversion ... do.	81.2	85.6	80.7	90.7	81.7	74.9	89.6	98.2	90.0
Hydrogen:carbon monoxide ratio:									
Fresh feed	2.93	2.73	2.92	2.95	2.98	2.94	2.95	2.93	2.92
Recycle, main reactor	54.5	29.0	30.4	-	35.6	45.3	-	-	-
Usage	2.24	2.25	2.24	2.60	2.31	2.02	2.56	2.86	2.55
Water vapor, recycle main reactor, volume-percent	9.5	10.2	9.4	9.4	9.3	13.3	13.2	13.3	12.8
Product gas, volume-percent (dry basis):									
H_2	65.4	57.9	63.9	39.2	60.5	67.9	45.0	8.0	43.7
CO	1.2	2.0	2.1	0.6	1.7	1.5	0.6	0.1	0.6
N_2	1.3	1.7	1.2	1.4	0.9	1.0	1.8	1.6	1.7
CO_2	3.7	5.5	3.7	0.0	3.8	7.2	0.0	0.1	0.0
C_1	16.8	19.2	17.6	45.7	20.2	13.1	37.3	89.6	40.3
$C_2{=}$	0.5	0.6	0.2	0.0	0.1	0.4	0.0	0.0	0.0
C_2	4.6	5.5	5.1	7.9	5.8	3.7	7.2	0.4	6.5
$C_3{=}$	2.4	2.8	1.9	0.3	1.6	1.9	0.4	0.1	0.3
C_3	2.0	2.3	2.4	3.9	2.9	1.6	5.6	0.1	4.9
$C_4{=}$	1.1	1.3	1.0	0.2	1.0	0.9	0.1	0.0	0.1
C_4	0.6	0.7	0.6	0.7	0.9	0.4	1.8	0.0	1.9
$C_5{=}$	0.3	0.4	0.2	0.0	0.3	0.3	0.0	0.0	0.0
C_5	0.1	0.1	0.1	0.1	0.3	0.1	0.2	0.0	0.0
$C_6{=}$	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
H_2O	9.5	10.2	9.4	17.7	9.3	13.3	30.4	31.6	32.5
Heating value product gas ... B.t.u./ft.^{3} [1]	654	705	656	876	709	573	881	946	870
Volume, fresh feed:volume, product gas	3.55	4.19	3.43	4.29	3.42	2.78	4.41	4.41	4.44
Yield, g./m.^{3} H_2+CO conversion:									
C_1	41.8	37.6	45.9	84.5	52.2	44.4	67.9	148.5	72.6
$C_2{=}$	2.2	2.1	0.9	0.0	0.4	2.4	0.0	0.0	0.0
C_2	21.7	20.8	25.2	27.6	28.3	24.5	24.8	1.3	22.2
$C_3{=}$	15.9	14.9	13.2	1.5	11.0	17.5	2.0	0.5	1.4
C_3	14.0	13.0	17.6	20.3	21.1	15.6	28.7	0.5	24.9
$C_4{=}$	9.9	9.3	9.4	1.4	9.3	11.2	0.6	0.0	0.6
C_4	5.6	5.3	5.9	4.9	8.7	5.2	12.4	0.0	13.0
$C_5{=}$	3.4	3.7	2.3	0.0	3.5	4.8	0.0	0.0	0.0
C_5	1.2	0.9	1.2	0.9	3.6	1.6	1.7	0.0	0.0
$C_6{=}$	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Oil	48.6	50.8	46.5	36.1	44.1	46.6	34.6	23.9	35.3
Aqueous phase	241.5	236.3	240.4	229.5	234.4	226.9	231.9	213.7	229.6
C_1-C_3OH [2]	10.8	9.6	10.4	7.9	7.6	11.0	4.3	5.3	1.9
Other oxygenates [3]	1.3	1.5	1.7	1.3	1.1	1.8	0.0	1.3	0.6
H_2O	229.4	225.2	228.3	220.3	225.7	214.1	227.6	207.1	227.1
CO_2	23.0	28.1	24.1	0.0	22.2	66.1	0.0	0.0	0.0
Hydrocarbon recovery ... g./m.^{3} H_2+CO conversion	176.4	169.5	180.2	186.4	190.9	186.6	177.0	181.3	172.5
Theoretical recovery ... g./m.^{3} H_2+CO conversion	198.5	199.1	197.1	190.4	195.1	197.7	194.9	183.7	192.3

See footnotes at end of table.

TABLE 3. - Experiment 21, hot-gas-recycle system (Con.)

Period	J	K	L	M	N	O
Hours of synthesis	1,019-1,091	1,163-1,211	1,219-1,267	1,307-1,355	1,403-1,479	1,487-1,535
Number of reactors	1	2	2	1	2	1
Main reactor:						
Space velocity ... hr.$^{-1}$	1,000	999	1,002	1,001	1,002	1,004
Recycle:fresh feed	21	18	20	20	20	20
Cold recycle:fresh feed	0.5	0.5	0.5	0.0	0.0	2.0
Pressure ... p.s.i.g.	400	400	402	401	402	402
Temperature, ° C.:						
In gas	300	300	300	300	300	300
Out gas	340	339	340	340	340	340
ΔT	40	39	40	40	40	40
Average	315	313	314	314	314	314
Pressure drop ... inches H_2O per foot	10	10	11	11	9	12
Second reactor, average temperature ... ° C.	-	345	363	-	365	-
Carbon dioxide-free contraction ... percent	59.6	76.2	76.7	51.2	75.3	68.7
Hydrogen conversion ... do.	58.0	96.2	98.5	46.1	92.5	73.1
Carbon monoxide conversion ... do.	95.5	99.3	99.7	91.5	99.1	94.4
Hydrogen + carbon monoxide conversion ... do.	67.6	97.0	98.8	57.7	94.2	78.5
Hydrogen:carbon monoxide ratio:						
Fresh feed	2.93	2.95	2.93	2.90	2.96	2.95
Recycle, main reactor	27.6	-	-	18.4	-	14.2
Usage	1.78	2.86	2.90	1.46	2.77	2.28
Water vapor, recycle main reactor, volume-percent	15.0	14.4	14.3	17.5	17.5	8.5
Product gas, volume-percent (dry basis):						
H_2	69.0	11.8	4.7	71.9	22.2	59.8
CO	2.5	0.7	0.3	3.9	0.9	4.2
N_2	0.8	1.4	0.8	0.6	1.4	1.0
CO_2	10.4	0.7	1.0	12.0	2.1	6.2
C_1	10.1	81.4	91.0	6.8	66.1	19.3
$C_2=$	0.4	0.0	0.0	0.3	0.0	0.0
C_2	2.6	2.2	1.3	1.6	3.8	4.5
$C_3=$	1.4	0.1	0.2	1.1	0.2	0.7
C_3	1.1	1.2	0.5	0.7	2.3	2.6
$C_4=$	0.8	0.1	0.0	0.6	0.1	0.5
C_4	0.4	0.4	0.2	0.2	0.8	0.8
$C_5=$	0.3	0.0	0.0	0.2	0.0	0.2
C_5	0.1	0.0	0.0	0.1	0.1	0.2
C_6	0.1	0.0	0.0	0.0	0.0	0.0
H_2O	15.0	41.7	41.9	17.5	47.9	8.5
Heating value product gas ... B.t.u./ft.^{3} [1]	507	955	986	429	839	1026
Volume, fresh feed:volume, product gas	2.22	4.18	4.26	1.80	3.97	2.99
Yield, g./m.^{3} H_2+CO conversion:						
C_1	47.7	144.4	155.5	47.2	127.4	59.0
$C_2=$	3.4	0.0	0.0	3.6	0.0	0.0
C_2	23.8	7.4	4.2	21.0	13.9	26.0
$C_3=$	18.0	0.5	0.9	20.4	1.0	5.7
C_3	15.0	6.0	2.4	13.6	12.5	22.4
$C_4=$	13.9	0.6	0.0	15.0	0.7	5.5
C_4	7.3	2.7	1.3	5.2	5.8	9.2
$C_5=$	6.6	0.0	0.0	6.4	0.0	2.7
C_5	2.2	0.0	0.0	3.2	0.9	2.9
$C_6=$	2.6	0.0	0.0	0.0	0.0	0.0
Oil	39.8	14.1	14.0	35.9	12.2	33.2
Aqueous phase	199.5	208.7	206.4	164.1	199.1	222.7
C_1-C_3OH[2]	6.1	0.6	2.0	9.0	0.2	7.8
Other oxygenates[2]	1.3	0.1	0.1	1.5	0.1	1.4
H_2O	192.1	208.0	204.3	153.5	198.8	213.6
CO_2	135.7	3.4	2.7	226.2	9.0	49.8
Hydrocarbon recovery ... g./m.^{3} H_2+CO conversion	187.7	176.4	180.3	182.0	174.7	175.8
Theoretical recovery ... g./m.^{3} H_2+CO conversion	197.1	183.4	182.2	197.4	185.1	194.6

[1] Cubic foot corrected to 60° F. and 30 in. Hg, dry.

[2] Calculated as hydrocarbons.

TABLE 4. - Composition of product gas as a function of fresh feed composition, with a steel catalyst at 300° to 340° C., 4:1 cold recycle to fresh feed ratio and 90 percent conversion; experiment 18, main reactor

Period	P	E	K
H_2:CO ratio, fresh feed	2.46	2.71	2.95
Cold recycle:fresh feed	4.0	4.0	4.0
Space velocity .... $hr.^{-1}$	800	800	800
Heating value, product .... B.t.u./ft.^{3} [1]	868	846	810
Product gas, volume-percent (dry basis):			
C_1	38.8	40.5	43.6
C_2-C_5 paraffin	15.0	13.3	10.0
C_1+C_2	45.8	48.1	49.6
C_3-C_5 olefin	0.7	0.6	0.6
H_2	31.2	34.7	38.8
CO	2.2	1.8	1.3
CO_2	11.0	8.1	4.4
Oil .... g./m.^{3} H_2+CO converted	16.6	10.4	5.5
Oil, percent of hydrocarbons recovered	9.0	5.7	3.2
Volume, fresh feed:volume, product gas	3.74	3.55	3.46
Water vapor, recycle main reactor .... volume-percent	4.7	4.9	4.8

[1] Calculated at 60° F., 30 in. Hg, dry.

The oil production was low, amounting to 3 to 9 percent of the total hydrocarbons, and increased as the carbon monoxide content in the feed gas increased.

The product gas leaving the reactor approached equilibrium with respect to the water-gas-shift reaction,

$$CO + H_2O = CO_2 + H_2.$$

The calculated mass action constant averaged 29, which is the equilibrium value at 319° C.

Cold Recycle

A portion of the recycle gas, equal to 2 to 6 volumes of the total recycle of about 20 volumes in experiment 18, was cooled to condense water vapor and thus vary the concentration of water vapor in the reactor. Water vapor content affects usage ratio, synthesis-gas conversion, and carbon monoxide content of the product gas. Table 5 compares several periods in which the cold recycle ratio ranged from 2.2 to 6.1. Increasing the cold recycle decreased the water vapor and carbon dioxide content and increased the usage ratio and carbon monoxide content. Similar results were obtained in experiment 21 when the cold recycle was decreased to 0.5 and then eliminated entirely. As the ratio of cold recycle-to-fresh gas was decreased, catalyst activity declined.

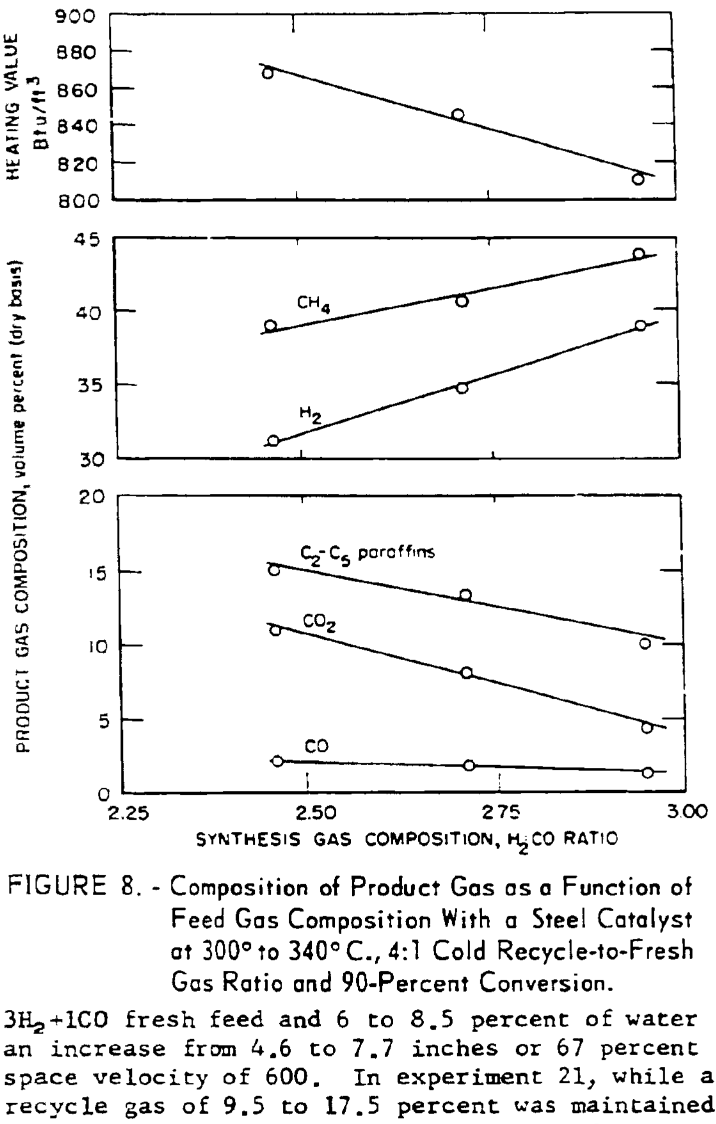

FIGURE 8. - Composition of Product Gas as a Function of Feed Gas Composition With a Steel Catalyst at 300° to 340° C., 4:1 Cold Recycle-to-Fresh Gas Ratio and 90-Percent Conversion.

Pressure Drop

There is a gradual increase in the pressure drop across the catalyst bed at temperatures above 300° C. However, operating with a high concentration of water vapor in the recycle gas decreases the rise in pressure drop significantly. The concentration of water vapor in the recycle gas is varied by changing the cold recycle gas flow.

In experiment 18, with a $2.7H_2+1CO$ fresh feed and a water vapor content in the recycle gas of 3 to 5 percent, the pressure drop increased from 13 inches of water per foot of catalyst to 30 inches or 130 percent in 300 hours of operation. This increase occurred even with a reduced flow of gas at the end of this period; that is, an hourly space velocity of 800, compared with an initial flow at 1,000 space velocity. In experiment 20, with a $3H_2+1CO$ fresh feed and 6 to 8.5 percent of water in the recycle gas, there was an increase from 4.6 to 7.7 inches or 67 percent in 550 hours at an hourly space velocity of 600. In experiment 21, while a water vapor content in the recycle gas of 9.5 to 17.5 percent was maintained, the pressure drop across the bed increased from 11 to 12 inches of water per foot of catalyst height, or 9 percent in 1500 hours of operation at an hourly space velocity of 1000 with a $3H_2+1CO$ fresh feed.

The effect of water vapor in retarding the increase in pressure drop is related to its ability to prevent carbon deposition and catalyst spalling. The lack of catalyst spalling is evident in the constant level of catalyst activity in experiment 21. The activity of the catalyst at the end of 1,500 hours of operation showed a very slight decline. At this time, period O, 78.5 percent conversion of synthesis gas was obtained compared with an initial conversion of 81.2 percent (period A)--both periods at identical operating conditions. Despite the large water vapor content the recycle gas was not sufficiently oxidizing to convert the iron carbide of the catalyst to the oxide.

TABLE 5. - Effect of varying the ratio of cold recycle to fresh gas, experiment 18, main reactor

Period	A	B	C
H_2:CO ratio, fresh feed	2.7	2.7	2.7
Cold recycle:fresh feed	2.2	4.0	6.1
Space velocity ..... hr.$^{-1}$	1,000	1,000	1,000
H_2+CO conversion ..... percent	84.2	85.9	86.7
Usage ratio, H_2:CO	2.18	2.28	2.36
Product gas, volume-percent (dry basis):			
CO_2	9.5	8.5	7.4
CO	2.0	2.8	3.2
CH_4	29.1	32.6	36.1
Water vapor, product gas ..... volume-percent	7.4	4.4	3.3
Heating value, product gas B.t.u./ft.^{3} [1]	721	771	768

[1] Calculated at 60° F., 30 in. Hg, dry.

Alkali Impregnation

Product distribution

Impregnating steel turnings with a small amount of potassium carbonate, equivalent to 0.02 percent K_2O, is sufficient to shift the product distribution to heavier hydrocarbons. This shift can be seen in table 6 in which a nonalkalized turning is compared with an alkalized turning. The methane yield dropped from 100.6 to 45.9 grams per cubic meter of H_2+CO converted, and the oil yield rose from 5.9 to 46.5 grams per cubic meter converted. In addition to the production of heavier hydrocarbons, the production of olefins and oxygenates increased significantly in the presence of alkali; C_2 to C_5 olefins increased from 4.9 to 25.8 grams per cubic meter converted and oxygenates from 1.2 to 12.1. Generally, little or no ethylene was produced. The production of olefins was further enlarged by increasing the water vapor content of the recycle gas. As shown in the same table, the C_2 to C_5 olefins increased from 25.8 to 45.4 grams per cubic meter converted as the cold recycle flow was decreased. Olefins increased despite the increase in hydrogen concentration of the recycle gas with an increase in water vapor content. Water vapor apparently inhibits the hydrogenation of olefins. Low molecular weight olefins are important for the production of alkylate gasoline. In period M, the C_2 to C_5 olefins accounted for 25 percent of the total hydrocarbon production.

Activity

Addition of alkali to the steel turnings moderately improved the catalytic activity. At the end of 500 hours of operation (experiment 21, period E) 81.7 percent conversion was obtained at an hourly space velocity of 1,000. The may be compared with a similar period under identical operating conditions with a nonalkalized turning (experiment 18, period J), which had 85.3 percent conversion at an hourly space velocity of 800. These activities when expressed as a product of space velocity and $\log_{10}$ (1/1-C)[18] are equivalent to 738 and 666, respectively--an increase of 11 percent. As the ratio of cold recycle to fresh gas is decreased, the activity of the catalyst decreases.

TABLE 6. - Effect of alkali impregnation on steel turnings and variation of cold recycle-to-fresh gas ratio on synthesis products

	Nonalkalized		Alkalized		
Experiment Period	18 J	21 C	21 F	21 J	21 M
H_2:CO ratio, fresh feed............	<------------------ 2.9 ------------>				
Cold recycle:fresh feed............	2.0	2.0	1.0	0.5	0
Water vapor, recycle main reactor, volume-percent....................	8.0	9.4	13.3	15.0	17.5
Yield, g./m.^{3} H_2+CO converted:					
CH_4..............................	100.6	45.9	44.4	47.7	47.2
C_2-C_6 olefins....................	4.9	25.8	35.9	44.5	45.4
C_2-C_6 paraffins..................	58.8	49.9	46.9	48.3	43.0
Oil................................	5.9	46.5	46.6	39.8	35.9
Oxygenates[1]......................	1.2	12.1	12.8	7.4	10.5
Total..........................	171.4	180.2	186.6	187.7	182.0

[1] Calculated as hydrocarbons.

Catalyst Composition

Steel turnings upon oxidation with steam form a coating of Fe_3O_4 and upon reduction, α-Fe. The depth of the layer of the 20-percent oxidized turnings was 0.002 inch. Figure 9 shows a cross section of the steel turning. The α-Fe is completely carbided to Hägg Fe_2C shortly after synthesis is begun, remaining as such throughout the run. Upon terminating the run, fines containing some of the catalyst coating, together with free carbon, were found at the bottom of the reactor. However, in experiment 21, operated with the high water vapor in the recycle gas, the quantity of fines was very small.

Single Reactor with Nickel-Turning Catalyst

When nickel turnings were substituted for steel turnings in the first reactor, experiment 19, a much lower activity resulted. A conversion of only 53 percent of a $3H_2$+1CO gas at an hourly space velocity of 550 and at an average temperature of 317° C. was obtained. Gradually increasing the temperature to 390° C. did not improve the activity.

Two Reactors--Raney Nickel Following Steel Turnings

By reacting the tail gas from the main reactor over a partially extracted Raney nickel in the second reactor, the conversion of synthesis gas increased

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[18] Anderson, R. B., Seligman, B., Shultz, J. F., Kelly, R., and Elliott, M. A., Fischer-Tropsch Synthesis, Some Important Variables of the Synthesis on Iron Catalysts: Ind. Eng. Chem., vol. 44, 1952, pp. 391-397.

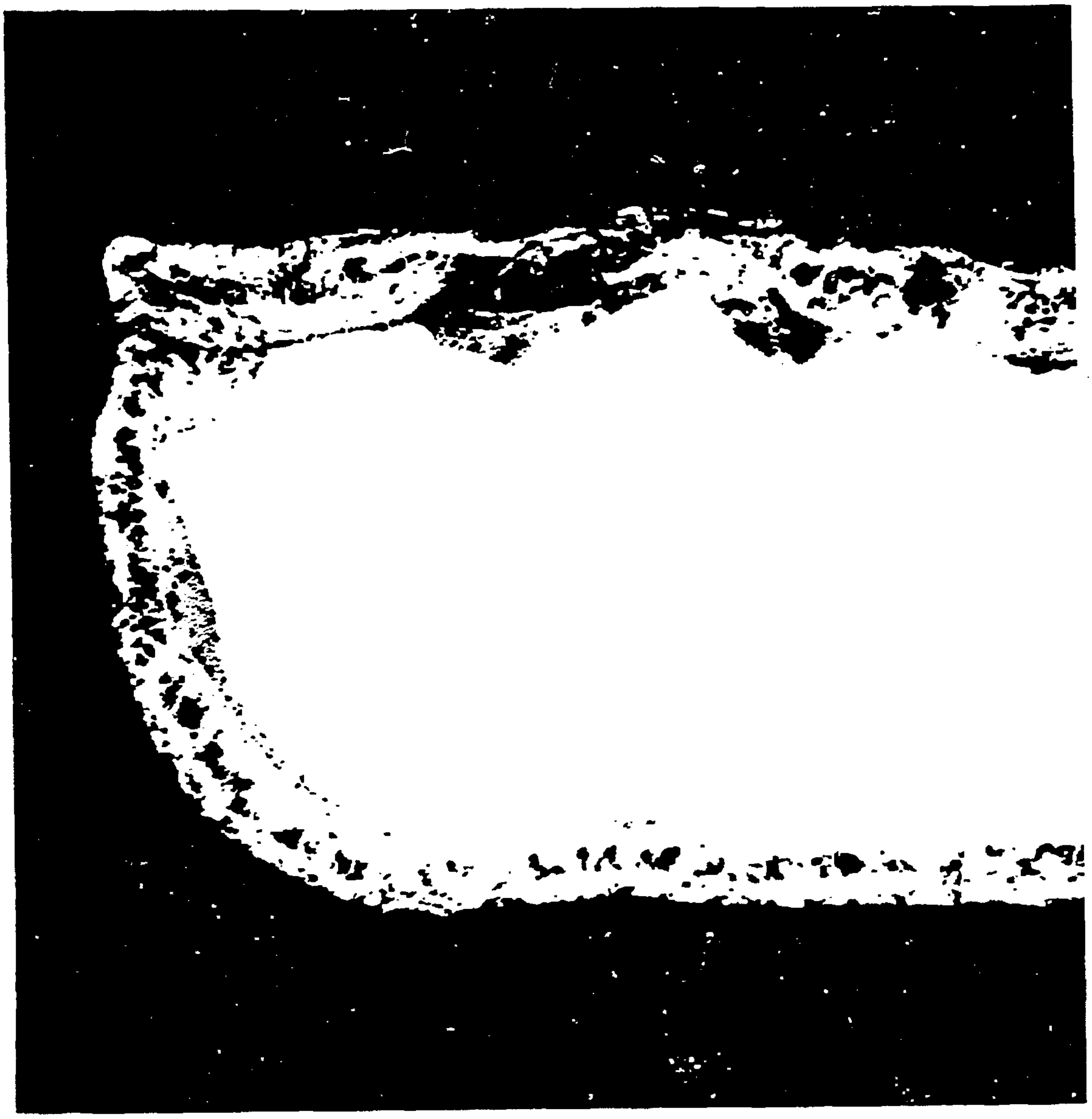

FIGURE 9. - Cross Section of Steel Turnings Catalyst. (X125)

to 99 percent and higher. Carbon monoxide practically disappeared, most of the carbon dioxide reacted, higher hydrocarbons and most of the unsaturated hydrocarbons were cracked and hydrogenated to methane. The heating value of the product gas increased to 950 B.t.u. and higher per cubic foot. The desired effects were obtained in all three experiments, 18, 20, and 21. Figures 10 and 11 show the change in composition of the product gas using a $3H_2$+1CO and $2.7H_2$+1CO fresh feed and operating the Raney nickel at 270°, 300°, and 350° C. With a $3H_2+1CO$ feed gas to the first reactor and Raney nickel at 350° C. in the second, the product gas contained 96 percent methane and had a heating value of 983 B.t.u. per cubic foot; with a $2.7H_2+1CO$ feed, 78 percent methane having a heating value of 1,130 B.t.u. was made. The higher heating value obtained with the 2.7 gas was due to the presence of about 14 percent of the higher hydrocarbons--ethane, propane, and butane. More fresh feed, however, is required to make the higher B.t.u. gas--4.8 volumes compared with 4.1. The complete composition of the product gases is shown in table 7.

TABLE 7. - Composition of product gas obtained with a $2.7H_2+1CO$ and $3H_2+1CO$ fresh feed using an iron catalyst at 300° to 340° C. followed by Raney nickel at 350° C., experiment 20

Period	M	H
H_2:CO ratio, fresh feed	2.75	3.02
H_2+CO conversion ... percent	99.7	99.9
Product gas, volume-percent, (dry basis):		
CH_4	78.2	96.0
H_2	1.2	0.6
CO	0.1	-
CO_2	3.7	1.8
C_2-C_5 paraffin	14.4	0.4
C_2-C_5 olefin	0.6	-
N_2	1.8	1.2
Heating value product gas ... B.t.u./ft.^{3} [1]	1,130	983
Volume of fresh feed:volume of product gas	4.82	4.09

[1] Calculated at 60° F., 30 in. Hg, dry.

Conversion of carbon dioxide by reduction,

$$CO_2 + 4H_2 = CH_4 + 2H_2O,$$

over the Raney nickel was high. As shown in table 8, about 90 to 95 percent of the carbon dioxide was converted with the $3H_2+1CO$ fresh feed and 70 to 85 percent with the $2.7H_2+1CO$ gas in the temperature range 270° to 350° C. In periods E and J only the first reactor, containing steel turnings, was operated. For both H_2:CO ratios, conversion of carbon dioxide was maximum at 300° C. In a test at 400° to 450° C. only about 60 and 20 percent of the carbon dioxide reacted with the $3H_2+1CO$ and $2.7H_2+1CO$ feed gases, respectively. The reduction of carbon dioxide to methane is highly desirable as it consumes a diluent of no heating value and excess hydrogen of low heating value, 325 B.t.u. per cubic foot. Another advantage of consuming carbon dioxide is that the need for carbon dioxide removal after methanation may be eliminated.

Nickel turnings were considerably less active when substituted for the Raney nickel in the second reactor. In period N, experiment 20, at 350° C., 91 percent conversion of the $2.7H_2+1CO$ fresh feed was obtained, producing a gas with a methane content of 47 percent and heating value of 811 B.t.u. per cubic foot. Temperatures above 350° C. may be necessary for satisfactory operation with the less-active nickel turnings.

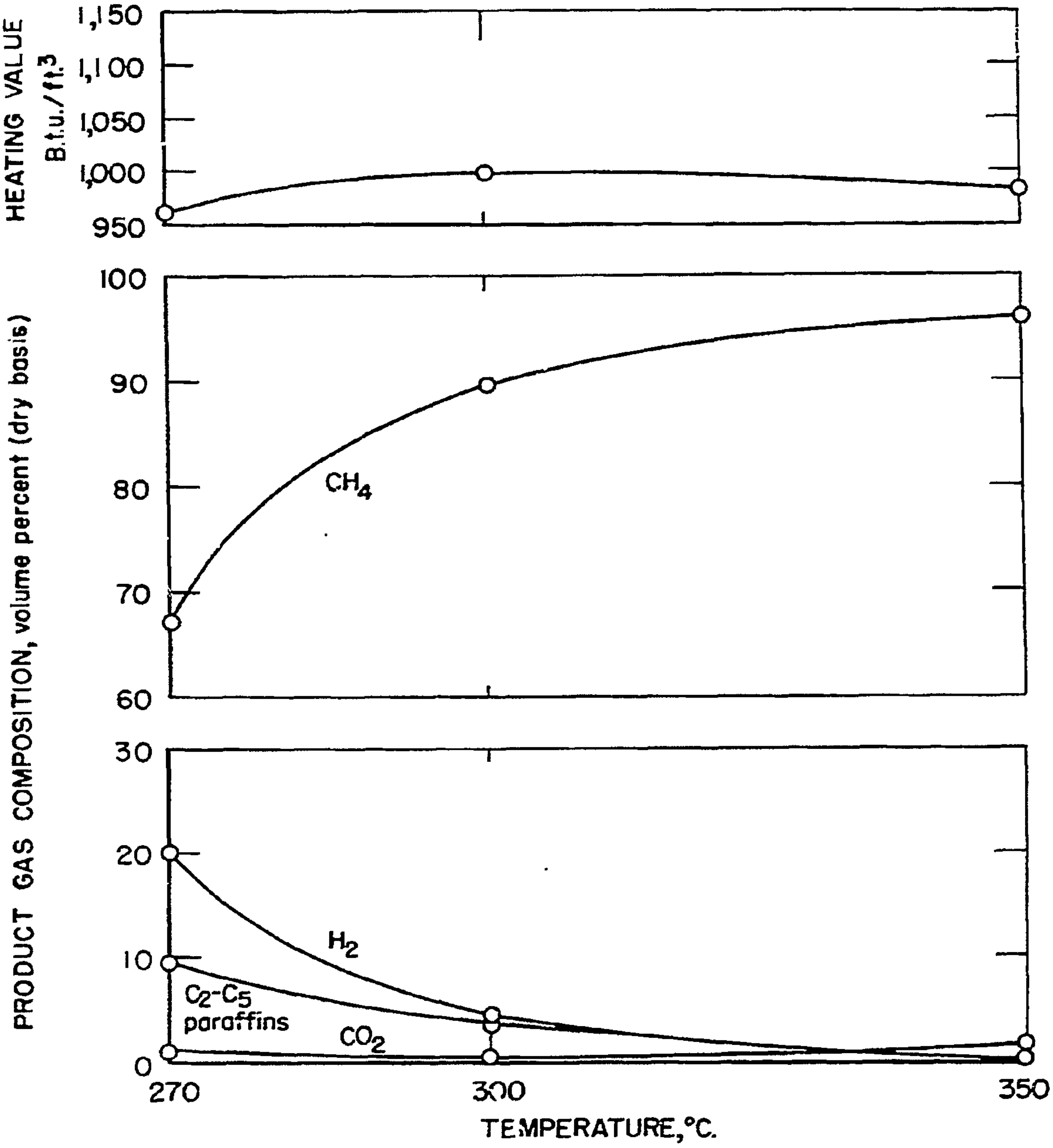

FIGURE 10. - Effect of Raney Nickel on Product Gas Composition Using a $3H_2$+1CO Feed Gas With a Steel Catalyst in Main Reactor at 300° to 340° C. and 2:1 Cold Recycle.

Table 9 shows the composition of Raney nickel. When discharged after 1,000 hours of operation, there was no evidence of fines. A nickel balance based on the analyses and weights of the charged and discharged catalyst indicated a loss of 7.2 percent nickel.

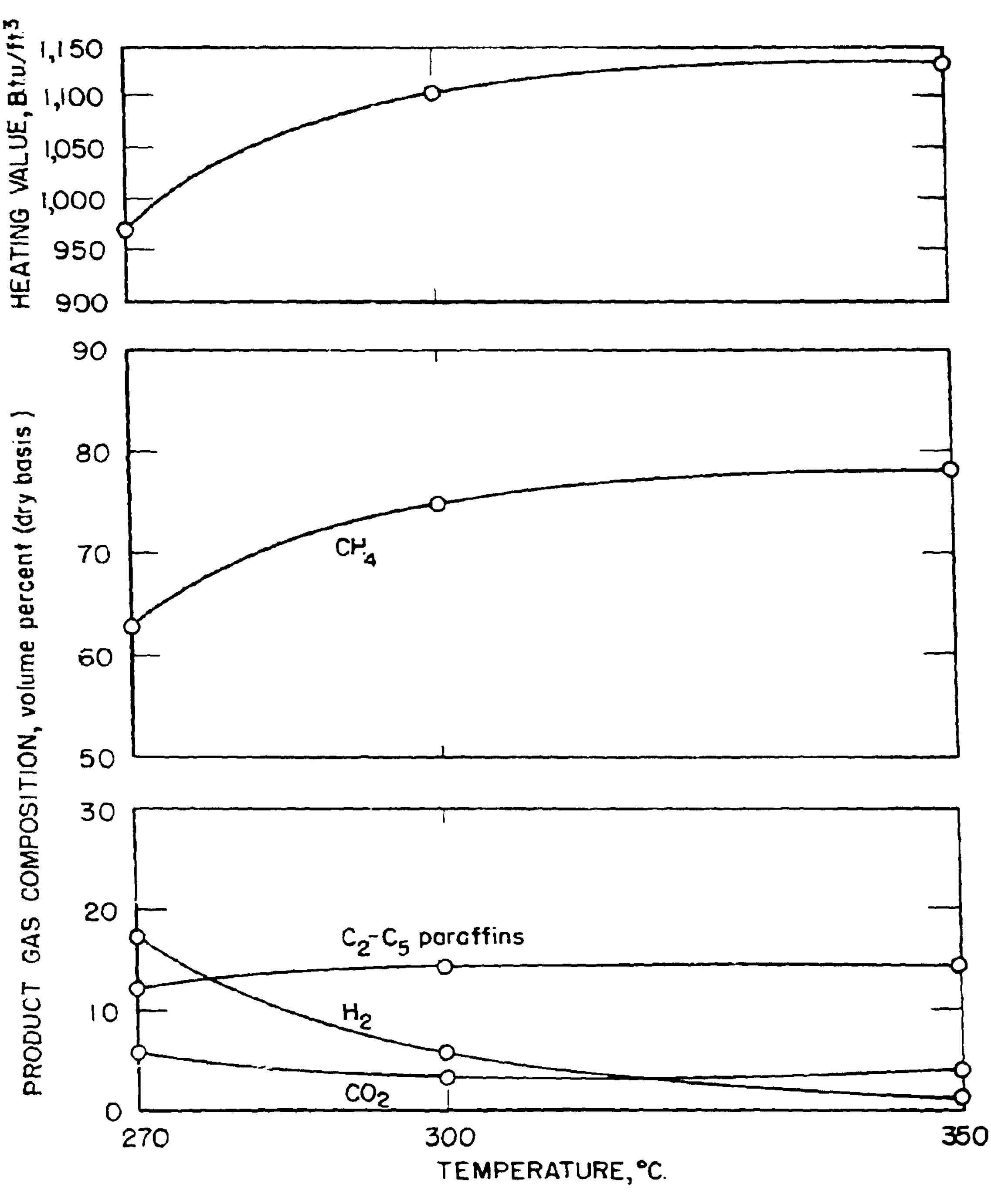

FIGURE 11. - Effect of Raney Nickel on Product Gas Composition Using a $2.7H_2+1CO$ Feed Gas With a Steel Catalyst in Main Reactor at 300° to 340° C. and 2:1 Cold Recycle.

TABLE 8. - Hydrogenation of carbon dioxide over Raney nickel, experiment 20, main reactor at 300° to 340° C., 2:1 cold recycle-to-fresh feed ratio

Period	H_2:CO ratio, fresh feed	Number of reactors	Temperature of 2d reactor, ° C.	CO_2 in product gas	
				Percent	Ft.^{3}/hr.
E.........	3	1	-	8.9	11.65
G.........	3	2	270	1.1	.72
F.........	3	2	300	.6	.38
H.........	3	2	350	1.8	1.21
J.........	2.7	1	-	11.0	11.79
K.........	2.7	2	270	5.8	3.69
L.........	2.7	2	300	3.3	1.87
M.........	2.7	2	350	3.7	2.12

TABLE 9. - Composition of Raney nickel catalyst used in second reactor

	Ni	Al	S	Na	C	Phase by X-ray analysis
	Weight-percent					
Raw, before leaching.......	41.4	59.7	0.035	-	-	Al_3Ni, Al, Al_3Ni_2
Charged to reactor after leaching..................	51.9	38.9	-	0.14	-	Ni, Al_3Ni_2, Al_3Ni, $Al_2O_3 \cdot 3H_2O$
Discharged, 100 hours of operation.................	49.5	40.8	-	<.05	0.35	Ni, Al_3Ni, Al

CONCLUSIONS

Use of the hot-gas-recycle system for synthesis of a high-B.t.u. gas has been demonstrated to be feasible in the pilot plant using a 3-inch by 10-foot reactor. In a single reactor, conversions of 85 to 90 percent of the synthesis gas, a mixture of 2.5 to $3H_2$+1CO, at an hourly space velocity of 800 to 1,000, at 300° to 340° C. have been obtained with a nonalkalized steel lathe turning as catalyst. Heating values of the product gas, on a dry basis, ranged from 810 to 868 B.t.u. per cubic foot, and were maximum with 2.5 gas. On a CO_2-free basis heating values ranged from 847 to 975 B.t.u. per cubic foot. Oil production was low, about 3 to 9 percent of the total hydrocarbons, increasing as the carbon monoxide content of the feed increased. Excellent temperature control, with no hot spots developing, was obtained with a 40° C. temperature differential in the bed and a total recycle of 20 volumes to 1 volume of fresh feed.

Impregnating the steel turnings with potassium carbonate shifted the product distribution to heavier hydrocarbons. The methane production decreased, whereas that of olefins and oxygenates rose sharply. Increasing the water vapor content of the recycle gas to 15 to 17 percent by decreasing the ratio of cold recycle to fresh feed to 0.5 or less increased the C_2 to C_5 olefin production to 25 percent of the total hydrocarbon yield. The use of a catalyst with a small amount of alkali may be of interest if byproduct materials, especially C_3 and C_4 olefins, are desired in addition to high-B.t.u. gas.

Maintaining a water vapor content in the recycle gas of more than 10 percent, decreased the tendency of carbon deposition and catalyst spalling. At the end of 1,500 hours of operation and an hourly space velocity of 1,000 there was an increase of only 9 percent in pressure drop.

With a second reactor containing Raney nickel at 300° to 350° C., the overall conversion of H_2+CO was increased to more than 99 percent. Product gas with heating values of 983 and 1,130 B.t.u. per cubic foot and containing 96 and 78 percent methane was obtained with a $3H_2$+1CO and $2.7H_2$+1CO feed gas, respectively. The gas contained virtually no carbon monoxide, 0.1 percent or less, and only a small concentration of unsaturated hydrocarbons, meeting all the present requirements of a high-B.t.u. gas. The advantages of using a steel catalyst for the bulk of the synthesis and a nickel catalyst for the remainder of the synthesis are twofold: (1) Catalyst cost is reduced and (2) the steel protects the nickel from sulfur poisoning by removing the sulfur. By decreasing the load on the nickel catalyst, high conversions are achieved at moderate temperatures, 300° to 350° C., which probably prolong the life of the nickel. No loss of activity was observed, and little nickel was lost after 1,000 hours on stream in intermittent duty in an experiment that lasted over 2,300 hours.

Further investigation of the hot-gas-recycle system is planned. A study of heat transfer and temperature control in a reactor of larger diameter, 12 inches, will provide data for extrapolation to units of commercial size.

END

POROUS STAINLESS STEEL FILTERS FOR REMOVING DUST FROM HOT GASES

By L. J. Kane, G. E. Chidester, E. Takach,
and C. C. Shale

report of investigations 5842

MARCH 3, 1849

UNITED STATES DEPARTMENT OF THE INTERIOR
Stewart L. Udall, Secretary

BUREAU OF MINES
Marling J. Ankeny, Director

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ILLUSTRATIONS (Con.)

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POROUS STAINLESS STEEL FILTERS FOR REMOVING DUST FROM HOT GASES[1]

by

L. J. Kane,[2] G. E. Chidester,[3] E. Takach,[4] and C. C. Shale[2]

SUMMARY

Several processes being developed by the Federal Bureau of Mines to provide new and more efficient uses for coal require practical methods for removing dust from hot gases. Filters provide such means. The efficiency of filters made of aluminum silicate fibers and porous stainless steel was determined. The results obtained with the fibers were previously reported.[5] In this investigation tests were made with three porosities of stainless steel filter material (grades C, D, and E) at a gas flow of 1,000 std. c.f.h./sq. ft. of filter, temperatures of 75° and 1,000° F., and dust loadings (as represented by cumulative dust fed) of 100, 1,000, and 10,000 gr./sq. ft. of filter. Pressure drop during the runs ranged from 0.1 to 21 in. of mercury, and dust removal from 25 to 99.93 pct.

Filters having the finest pores gave the greatest pressure drop and percentage of dust removal; however, their dust-holding capacity was slightly lower. Usually, their ability to remove a higher percentage of dust at the same pressure drop more than compensated for the slightly lower dust-holding capacity.

With increase in temperature, for the same weight rate of flow of gas, the pressure drop increased and the percentage of dust removal decreased. The effect of temperature was probably due to the accompanying change in the linear velocity and viscosity of the gas.

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[1] Work on manuscript completed February 1961.

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[4] Former student trainee (electrical engineer), Morgantown Coal Research Center, Bureau of Mines, Morgantown, W. Va., now electrical engineer, Weirton Steel Co., Weirton, W. Va.

[5] Kane, L. J., Chidester, G. E., and Shale, C. C., Ceramic Fibers for Filtering Dust From Hot Gases: Bureau of Mines Rept. of Investigations 5672, 1960, 18 pp.

For the same cumulative dust fed, the concentration of the dust in the feed had no effect on the pressure drop or the percentage of dust removed. The higher the loading of the filter, the greater was the pressure drop and the percentage of dust removed.

Blowing clean gas at higher velocity back through the filter for a few seconds effectively removed dust from the filter. A second blast had no appreciable effect. After blowback, pressure drop and percentage of dust removed were slightly higher than the original values.

At equal pressure drop across the dust-laden filters, porous stainless steel filters were less efficient than aluminum silicate fibers. However, fiber filters could not be regenerated by reverse flow of gas.

Both types of filter can be used up to 1,000° F. Tests have shown that aluminum silicate fiber filters can be used effectively up to 1,800° F. A porous stainless steel filter appears practical for gases containing relatively high concentrations of dust, in which case it can be used where frequent regeneration is required.

INTRODUCTION

Removal of dust from gases at high temperatures is an important part of the Bureau's coal-gasification research and development program and an integral part of the coal-fired gas-turbine program.

Synthesis gas produced by the Bureau's coal-gasification processes contains dust and other contaminants which must be removed to avoid fouling catalysts and damaging chemical equipment used in gas-synthesis processes. Because the cost of purifying the gas is a substantial part of overall cost, purification must be accomplished most economically. If the dust can be removed without materially cooling the gas, heat exchange equipment can be eliminated and significant reduction in cost can be realized. In these processes, filters may be required at operating conditions of 300° to 800° F. If the heat for gasification is obtained from nuclear energy, it might be necessary to completely remove small quantities of dust from recycle helium at temperatures possibly as high as 2,500° F.

The Locomotive Development Committee (L.D.C.) of Bituminous Coal Research, Inc., which began development work on a coal-fired gas turbine that was later assigned to the Bureau at Morgantown, W. Va., found that erosion of turbine blades by dust in the hot gases (1,400° F. and 80 p.s.i.g.) was a big problem and removal of the dust quite difficult.[6] The L.D.C. developed a cyclone (Dunlab) separator to remove the dust, but enough remained to cause considerable damage.[7]

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[6] Yellott, J. I., Broadley, P. R., Myer, W. M., and Rotzler, P. M., Development of Pressurizing, Combustion, and Ash Separation Equipment for a Direct-Fired, Coal-Burning Gas Turbine Locomotive: ASME paper 54-A-201, November and December 1954, p. 14.

[7] Bituminous Coal Research, Inc., Report on 750 Hour Test of 4250 Horsepower Coal-fired Gas Turbine Power Plant: Dunkirk, N.Y., 1952, pp. 4-7.

Thus, the development of economical methods for removing dust at high temperature is important to the Bureau in improving some processes that utilize coal.

Bureau researchers have considered several types of dust-removal equipment for these applications. They found that the efficiency of cyclone separators is limited and that electrostatic precipitators that operate under the required conditions are not commercially available. For certain of these applications, however, filters might provide a practical solution. Filters can remove dust to almost any desired degree, provided they are deep enough or their cross-sectional area is great enough.

The Bureau has not investigated the use of bag filters. Although extensively applied in industry, and readily regenerated, they cannot be used above about 650° F. Filters of aluminum silicate fibers are satisfactory up to 1,800° F.,[8] but much of the dust is caught deep within the fibers, making regeneration by blowback nearly impossible. Thus, fiber-type filters appear practical only for gases containing extremely low concentrations of dust, when the filter can be used for a relatively long time without replacement.

Porous stainless steel filters appeared attractive for removing high concentrations of hot dust because they have uniform porosity, are rigid, operate up to 1,000° F., and offer the promise of satisfactory cleaning by blowback.[9] Thus, the filtering characteristics of porous stainless steel filters, such as change in pressure drop and percentage of dust removal, were studied over ranges of (1) dust loading on the filter, as represented by cumulative dust fed, (2) dust concentration in the feed gas, and (3) temperature. The results were then compared with those of the aluminum silicate fibers previously investigated.

APPARATUS AND PROCEDURE

The apparatus is shown in figure 1. The filter was a type-304 (18-8) porous stainless steel disk welded into a 1-3/16-in.-inside-diameter type-304 stainless steel tube. Three grades of filters, manufactured by the Micro Metallic Corp., Glen Cove, N.Y.,[10] were used (fig. 2). Characteristics of these filters are given in table 1.

TABLE 1. - Properties of porous stainless steel filter material tested

Grade	Thickness, inches	Mean pore openings, microns	Resistances to air flow of 1,000 std. c.f.h./sq. ft. c.s.a.[1] of filter at room temperature, in. Hg
C................	1/8	165	0.0135
D................	3/32	65	.034
E................	3/16	35	.273

[1] C.s.a. = cross-sectional area.

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[8] Work cited in footnote 5.

[9] Pall, D. B., Filtration of Fluid Catalyst Lines From Effluent Gases: Ind. Eng. Chem., vol. 45, 1953, pp. 1197-1202.

[10] Reference to specific makes and models of equipment is made to facilitate understanding and does not imply endorsement of such brands by the Bureau of Mines.

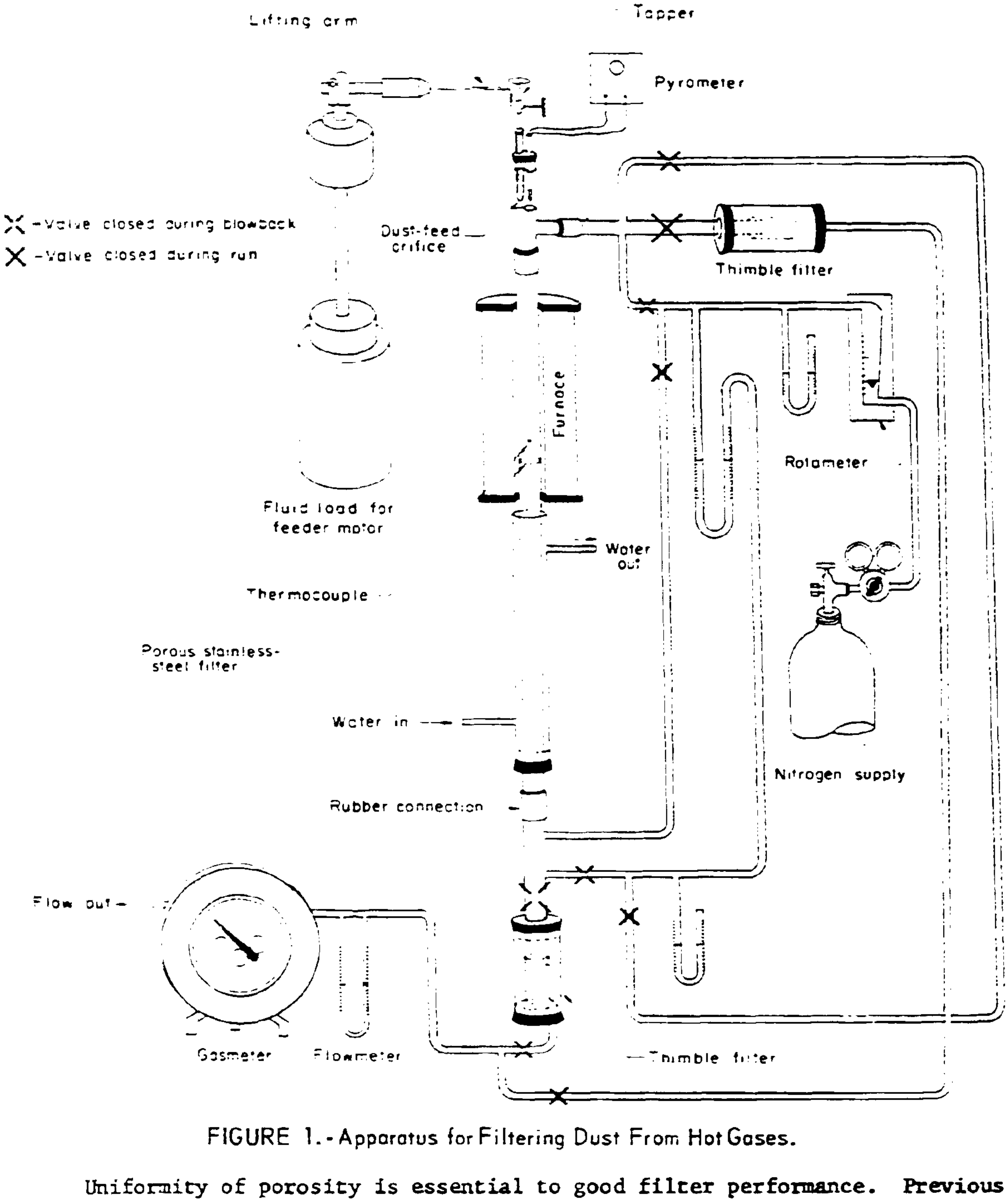

FIGURE 1.-Apparatus for Filtering Dust From Hot Gases.

Uniformity of porosity is essential to good filter performance. Previous work with fiber filters revealed that gas streams tend to channel through the more loosely packed areas, where percentage of removal is lower, and much of the filter is relatively unused. Observation with a hand lens showed that the

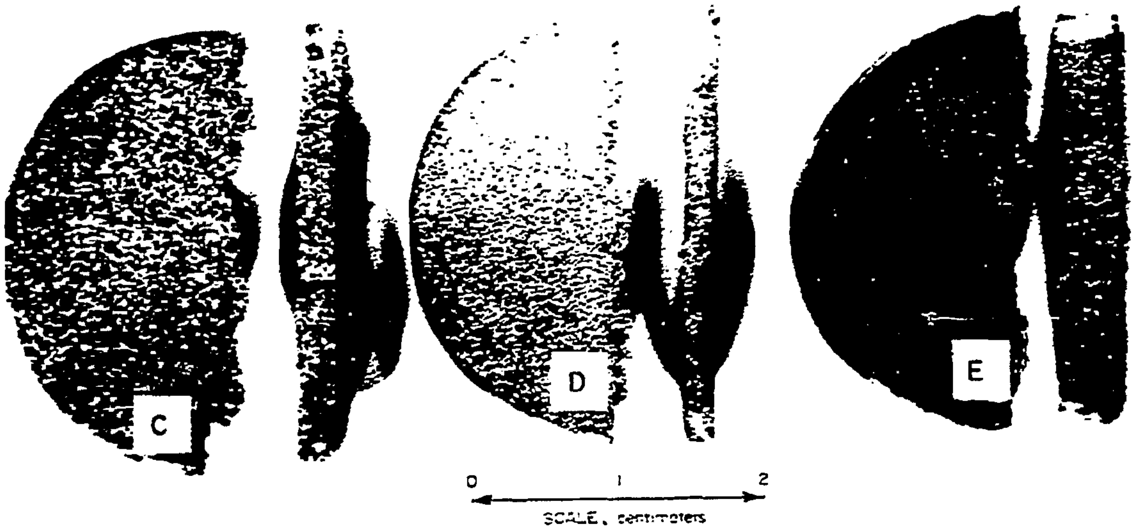

FIGURE 2. - Porous Stainless Steel Filter Disks, Grades C, D, and E.

porosity of the stainless steel filters was quite uniform, however. To prevent changes in pore size near the tube wall, it was necessary to avoid overheating the porous stainless steel disks when they were welded into the tube. Change in porosity was readily detected by observation and by comparing the flow resistance of the filter disk before and after it was welded. Use of electric welding was essential to avoid overheating the filter disk, and special care had to be taken to prevent splattering of metal on the surface of the filter.

Maximum temperature at which the material would operate without significant change in pressure drop was determined by measuring the pressure drop across clean disks with air at 75° F. and 1,000 std. c.f.h./sq. ft. of filter, before and after the disks were heated. Any increase in pressure drop was considered unsatisfactory. About 1,200° F. was the maximum temperature for grade C and 1,000° F. for grades D and E.

A measured quantity of dust was added to the feed gas by a vibrating feeder developed during previous Bureau research work.[11] Nitrogen was used as the carrier to reduce explosion and physiological hazards. The dust-laden gas was passed through the filter under the desired conditions. The gas leaving the filter was cooled by means of a water jacket surrounding the lower end of the tube; the dust remaining in the gas was filtered with a paper thimble and weighed.

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[11] Work cited in footnote 5.

FIGURE 3. - Fly Ash Used as Test Dust.

Figure 3 is a photomicrograph of the dust used in all tests. The particle-size distribution of this material was determined by dispersing the dust in water, rapidly filtering a drop through a membrane filter and counting under a microscope the number of particles within each particle size range.[12] A full analysis is given in table 2. The dust, most of which was less than 20 microns in size, was stored in air at 50 pct. relative humidity to facilitate feeding. Moist dust tends to pack, and dry dust sometimes tends to agglomerate due to electrification. The dust used in this work contained as much as 8 pct. moisture without appearing sticky. Approximately 5 pct. of the dust not retained by the filter adhered to the relatively cool stainless steel tube immediately downstream, compared with 2 pct. in the Vycor tube used in the investigation of fibers. Dust did not adhere in the heated zone, conforming with the thermal precipitation theory, which states that more dust will adhere to surfaces that are colder than the gas.

Because these were the initial tests on this filtering material, operating conditions were varied over a wide range (table 3). Table 3 also includes values that remained unchanged throughout the tests. Dependent variables that were investigated were pressure drop across the filter and percentage of dust removed from the gas.

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[12] Kane, L. J., Wright, H. C., and Shale, C. C., Use of Membrane Filters for Determining the Size of Dust Agglomerates as They Actually Exist in a Gas Stream: Bureau of Mines Rept. of Investigations 5637, 1960, p. 12.

TABLE 2. - Size distribution of test dust

Size of dust, microns	Percentage		Size of dust, microns	Percentage	
	By count	By volume		By count	By volume
>20...........	0.6	28.8	2-5............	27.8	1.8
10-20.........	12.1	62.3	0-2............	48.7	.1
5-10..........	10.8	7.0			

TABLE 3. - Materials and test conditions for investigation of porous stainless steel filters

Filter tube......................................	Type-304 stainless steel
Filter material..................................	Do.
Diameter of filter..............................in.	1-3/16
Grades...	C, D, and E
Dust...	Fly ash
Particle size of dust............................	Minus-325 mesh
Cumulative dust fed, gr./sq. ft. c.s.a.[1] of filter.	0, 100, 1,000, 10,000
Carrier gas......................................	Nitrogen
Gas flow, std. c.f.h./sq. ft. c.s.a. of filter.....	1,000
Concentration of dust in gas, gr./100 cu. ft......	100, 1,000
Temperature.................................° F.	75, 1,000

[1] C.s.a. = cross-sectional area.

EFFECT OF OPERATING VARIABLES ON PERCENTAGE OF DUST REMOVED AND PRESSURE DROP

Cumulative Dust Fed

The data points in graphs in which cumulative dust fed was the independent variable do not represent values at any instant, because it was necessary to determine the percentage of dust removed over a period of time, and hence over a range of dust fed, in order to obtain enough dust to weigh. In order to correspond to percentage-removal figures, pressure drop values also had to be averaged over the same range. Thus, the plotted values for pressure drop are averages.

Figure 4 shows that accumulation of dust on the filter greatly increased the pressure drop across grades D and E but had little effect on pressure drop across grade C, possibly because it was more difficult for dust to fill the larger pores of grade C, or because only the larger particles were retained. The fact that about 10 pinpoints of light could be seen through each square inch of clean grade-C material indicated that some dust could pass through without contacting the filter. After considerable dust-laden gas had been passed into the tube, a bed of dust accumulated on the filter. Slopes of the curves in figure 4 above 1,000 gr./sq. ft. were caused by the dust bed, while slopes below 1,000 were caused mainly by the porous stainless steel. Above 1,000 gr./sq. ft. the slopes for grades D and E were the same; below 1,000 the slope was lower for D, higher for E.

The effect of buildup of a dust bed on increase in pressure drop can be shown more clearly by graphing the increase caused by increase in concentration of dust on the filter, $\frac{d\,(\Delta p)}{d\,(DF)}$, against the cumulative dust fed (fig. 5).

(These differentials were calculated from actual pressure drops at the beginning and end of each of the three periods within each run, instead of from average values during each period as was done in figure 4. This could be done in this case because the percentage of removal, which must be an average value, was not involved.) Since it is desirable that the time between regenerations be as long as practicable, it is important that the pressure drop increase as little as possible with increase in dust load. The ratio, $\frac{d\ (\Delta p)}{d\ (DF)}$, is a measure of this factor in evaluating filters. Figure 5 shows that the average differential, in $\frac{\text{inches of mercury}}{\text{grains per square foot of cross sectional area}}$ was about 13×10^{-4} for grade E, 9×10^{-4} for grade D, and 10×10^{-4} for the dust bed. The value for grade C was always less than 3×10^{-4} and usually considerably less.

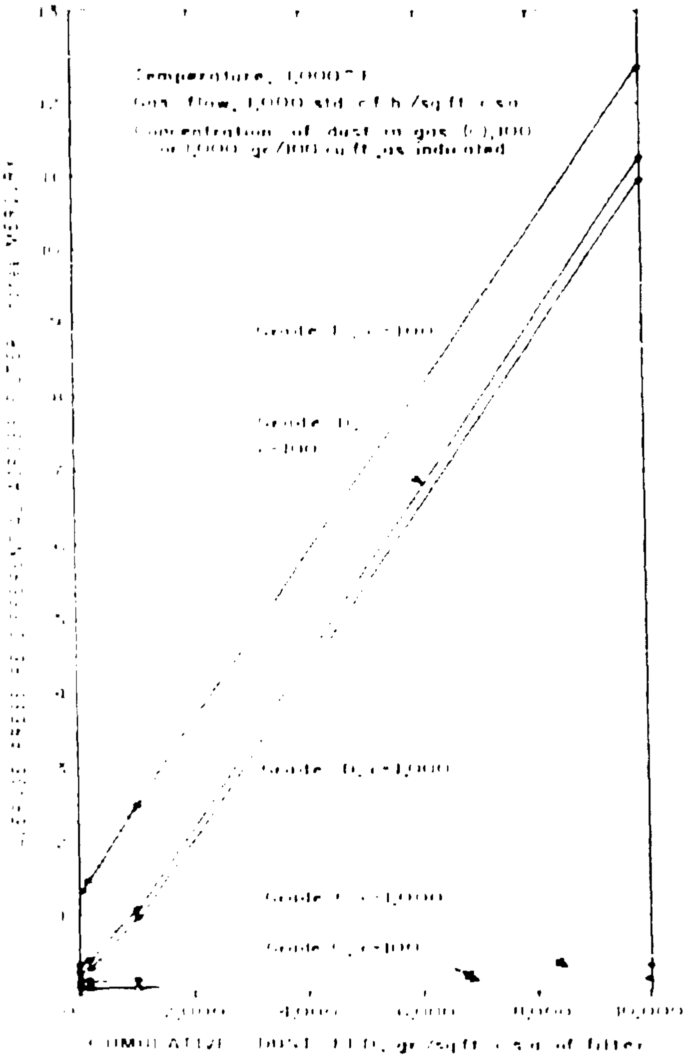

FIGURE 4. - Effect of Dust Fed and Concentration of Dust in Feed Gas on Pressure Drop Across Different Grades of Porous Stainless Steel Filters.

Another very important variable in dust filtration is the percentage of dust removed under various conditions. As shown graphically in figure 6, the ordinate of which is an inverse logarithmic scale, the percentage of dust removed from the gas rose as more dust was fed.

From a practical standpoint, the greatest possible removal of dust at a specified pressure drop is a most desirable quality for a filter. Figure 7 and 8 give a direct comparison of different filters on this basis. The same data are shown in both graphs, but in figure 7 the three points on each curve (lines of constant porosity) are results with 100, 1,000 and 10,000 grains of dust fed per square foot of cross-sectional area of filter, while in figure 8 the three points on each curve (lines of constant dust fed per square foot of filter) are results with the three grades of filtering material. These data were

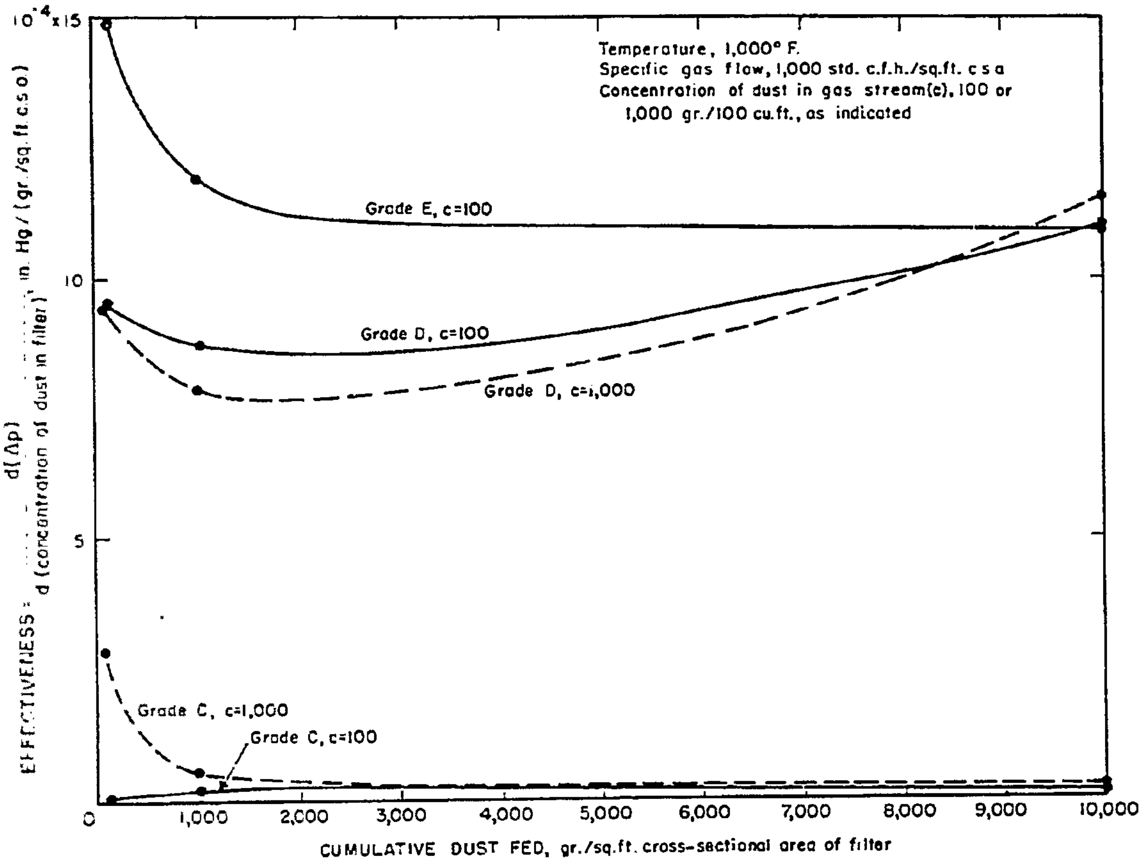

FIGURE 5. - Effectiveness of Different Grades of Filters as a Function of Dust Fed.

obtained with various concentrations of dust in the feed gas, but the effect of this variable was negligible, as shown later.

According to the impingement theory, very small particles of filter material, nearer the size of the dust particles, remove dust much more effectively than relatively larger particles, such as the stainless steel spheres. By comparison on this basis, porous stainless steel should remove dust less effectively than fibers. Figure 8 shows that this was found to be true. On the basis of the same theory, the bed of dust on the stainless steel filter should have given greater filter efficiency at the same pressure drop than the porous stainless steel itself because the particles of dust in the bed were about the same size as those in the gas. Figure 8, however, indicates the opposite to be true; higher concentrations of dust in the filter gave a lower percentage of removal at the same pressure drop. This effect was not noticeable with the aluminum silicate fibers, probably because a considerable quantity of the dust was caught within the bed instead of on or near the surface, as was the case with the stainless steel.

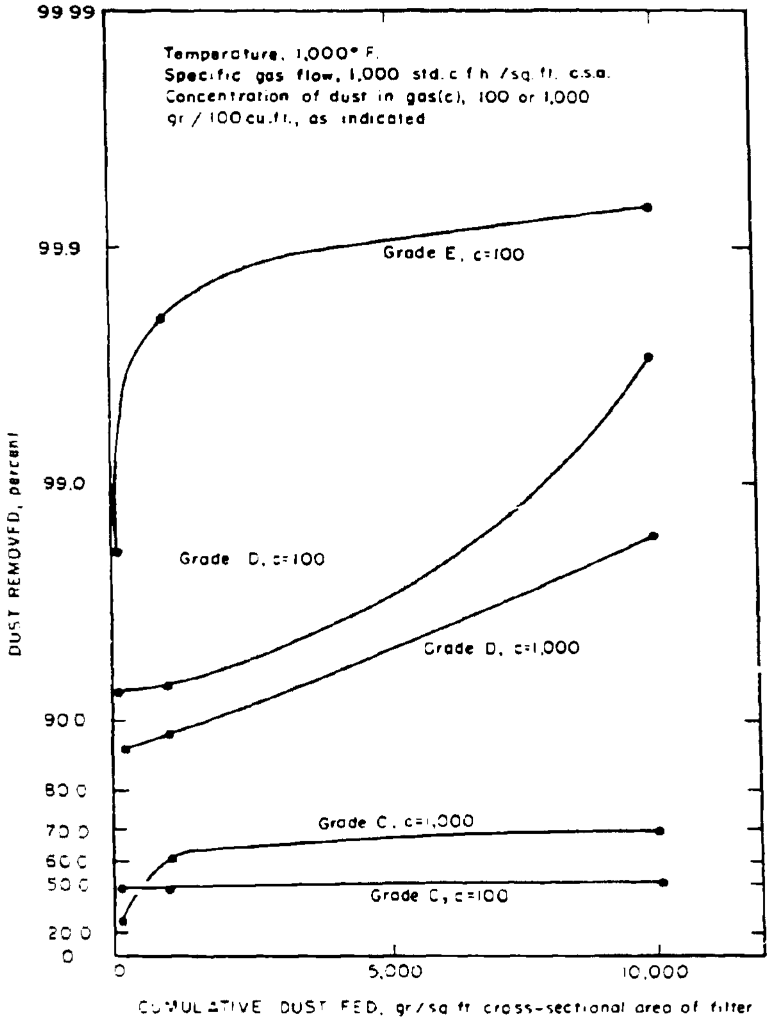

FIGURE 6. - Effect of Amount of Dust Fed and Concentration of Dust in Feed Gas on Percentage of Dust Removed by Different Grades of Porous Stainless Steel Filters.

The dust capacity of the filter should also be considered in this discussion. A filter that removes a high percentage of the dust at low pressure drop can be of little value if it plugs too rapidly. This effect may be seen by comparing the different sets of curves drawn through the same points to show the effects of porosity (fig. 7) and dust fed (fig. 8). A great increase in the percentage of dust that was removed usually more than compensated for a lower dust capacity of grade E stainless steel. For example, the grade D filter, after passage of about 2,000 gr. of dust per square foot of cross-sectional area, offered a resistance of 2 in. of mercury and removed about 94 pct. of the dust. The grade E filter reached the same pressure drop with only half this amount of dust passed, which means it would have to be regenerated twice as often, but removed 99.5 pct. of the dust. Under other conditions, however, grade D may be the most desirable. For instance, if 98.5 pct. removal is required, grade D will provide it with a pressure drop of 10-in. of mercury and about 9,000 gr. of dust fed per square foot of cross-sectional area. Before this time, the gas would need further treatment if the filter were completely cleaned by the previous regeneration. Grade E would require only 1.7 in. of mercury, but, with only about 200 gr. of dust fed per square foot of cross-sectional area, it would have to be regenerated frequently. Thus, this graph can be used for choosing the most desirable conditions for the required results. The dotted line shows the superior performance of the fiber filters--a much better percentage of removal at lower pressure drop.

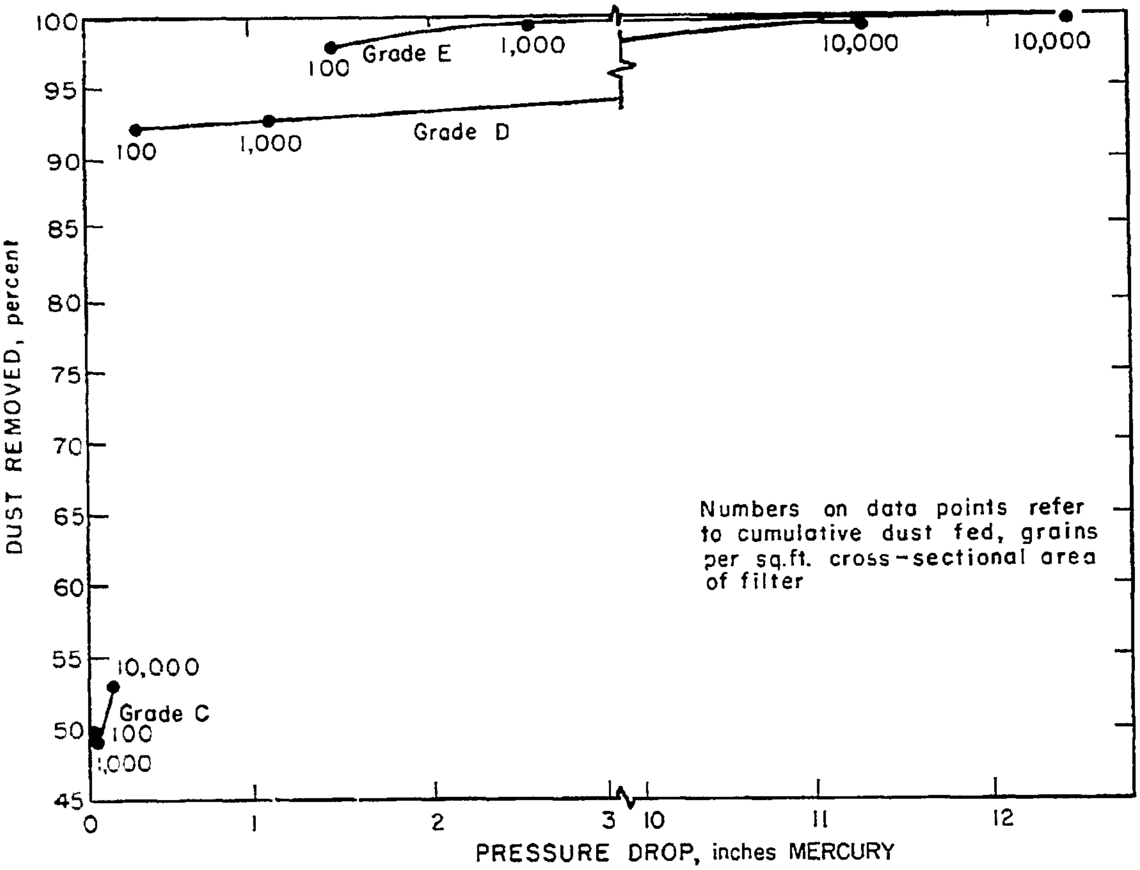

FIGURE 7. - Efficiency of Dust Removal as a Function of Pressure Drop Across Different Grades of Filters.

The relationship among these four practical variables--pressure drop, percentage of dust removed, dust capacity, and grade of filtering material--is shown more clearly for some purposes in figure 9. It is readily apparent that the large increase in percentage of removal that results from the substitution of grade E for grade D usually more than compensates for the slight decrease in capacity at the same pressure drop.

Resistance of Clean Filters

As would be expected, filters of higher initial resistance always gave higher pressure drop and percentage of dust removal, figures 4 and 6. This was also the case in the previous work on fiber filters. M. W. First, however, in the one comparison made, found that the more loosely packed fibers gave a somewhat higher percentage of removal.[13] First believed that this

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[13] First, M. W., Graham, J. B., Butler, G. M., Walworth, C. B., and Warren, R. P., High Temperature Dust Filtration: Ind. Eng. Chem., vol. 48, 1956, p. 700.

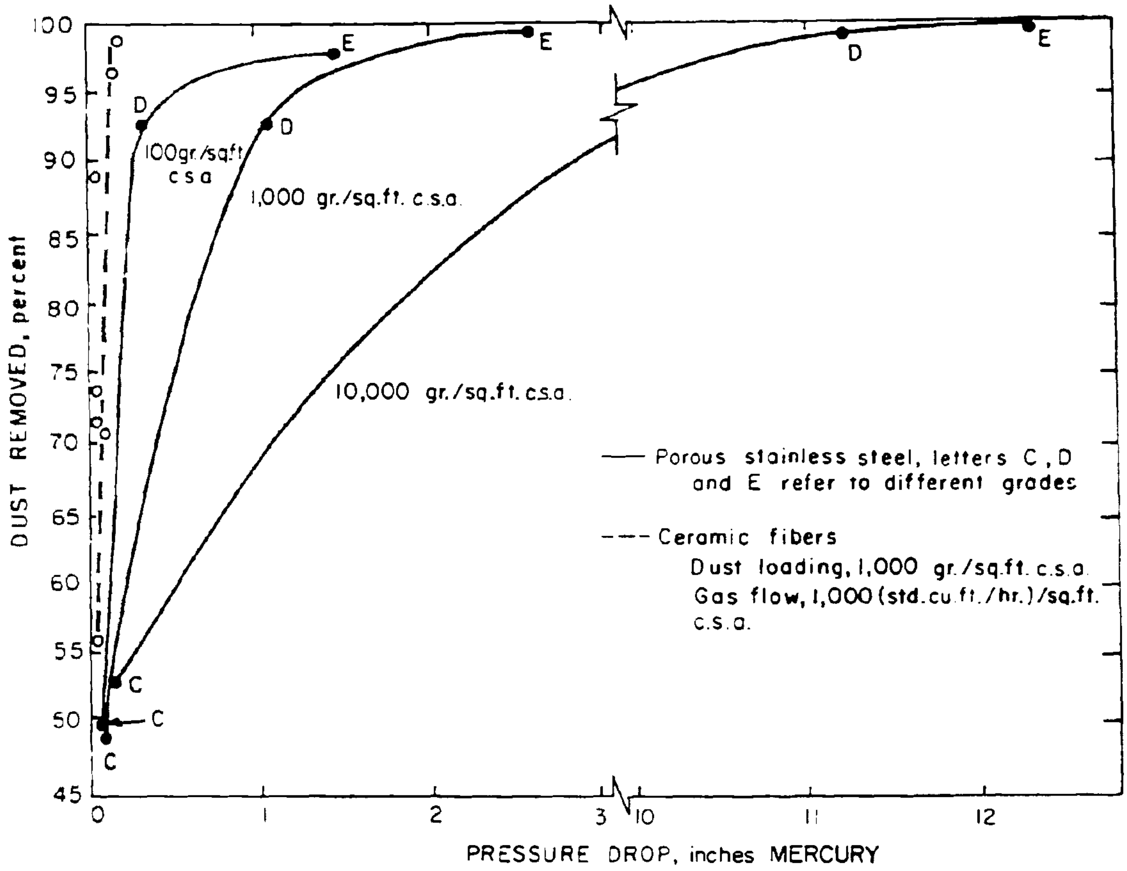

FIGURE 8.- Efficiency of Dust Removal as a Function of Pressure Drop Across Filters With Different Amounts of Dust Fed.

was due to the difference in fiber dispersion and uniformity of packing. Although the grade E filter had highest initial pressure drop, addition of large amounts of dust did not increase the pressure drop of E any more than that of D, while percentage of removal increased greatly (figs. 4 and 5). This makes grade E the most desirable filter for high dust loadings, provided relatively high pressure drop can be tolerated. Figure 9 shows that this sacrifice is not great. At low loadings, however, the pressure drop of grade E was several times that of grade D. Grade C always gave very low pressure drop and percentage of removal, neither of which changed much as dust was added to the filter. Thus, a loose filter is suitable for high loading, low pressure drop, and low performance. This was also true of fiber filters; hence, it seems to be a characteristic of loose filters and not caused by any property of the filtering material, such as nonuniformity of porosity.

Concentration of Dust in Gas

Figure 4 shows that the concentration of dust in the gas did not affect pressure drop. Curves for the grade D filters are close together and the

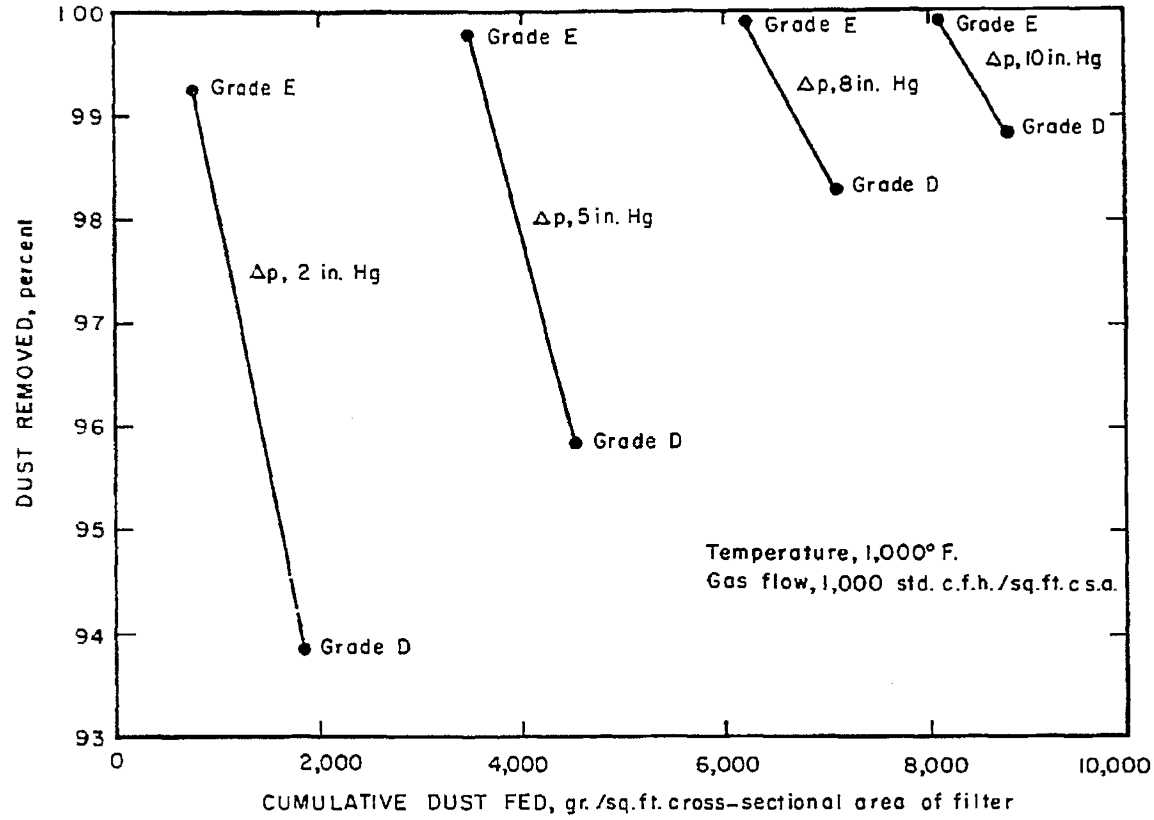

FIGURE 9. - Characteristics of Different Grades of Porous Stainless Steel Filters at the Same Pressure Drop.

same is true for grade C. Figure 6 shows that as the concentration of dust in the gas increased, the grade D filter removed slightly less dust, while grade C usually removed slightly more. These effects are negligible, however, compared with a ten fold increase in dust concentration in the gas and are probably caused by slight differences in the filters or dust samples. Rowley[14] obtained these same results with several different types of filters when filtering gases containing much lower concentrations of dust. As a standard when rating filters, the use of a gas containing a high concentration of dust was recommended because the results are the same and the work can be done more rapidly. Thus, the conclusions presented in this paper probably apply even when the concentration of dust in the gas is much lower--a more advantageous condition for using filters because they obviously would not have to be regenerated as often.

Temperature

For equal weight flow of gas in the viscous range, pressure drop across a filter of any specified porosity should be proportional to the change in volume (linear velocity) and viscosity of the gas with temperature. For air, the ratio of pressure drop at 1,000° F. to the pressure drop at 75° F. should be 6.0. The effect of flow on pressure drop (table 4) indicates that the flow was not viscous in any of the tests because pressure drop was not directly proportional to the first power of the flow rate. If flow is viscous, pressure drop should be directly proportional to flow. However, this effect may have been caused by entrance losses as the air entered the thin disk. Except when there was a large quantity of dust in the filter, the ratio of pressure drop at 1,000° F. to pressure drop at 75° F. was near the theoretical value of 6.0 (table 5). The last value in the table (2.9) indicates that the particles of dust in the dust bed were much coarser than the stainless steel particles in the filter--giving greater porosity, hence, greater departure from viscous flow--and corroborating the conclusion previously made by comparing the percentage of dust removed by the filters with that removed by the dust bed. This effect did not occur at 1,000° F.

TABLE 4. - Effect of flow on pressure drop

	Grade C		Grade D		Grade E	
Δp, in. Hg	Flow,[1] cu. ft./hr.-sq. ft.	Ratio of flow to Δp, cu. ft./hr.-sq. ft.-in. Hg	Flow[1] cu. ft./hr.-sq. ft.	Ratio of flow to Δp, cu. ft./hr.-sq. ft.-in. Hg	Flow,[1] cu. ft./hr.-sq. ft.	Ratio of flow to Δp, cu. ft./hr.-sq. ft.-in. Hg
0.02	240	12,000	78	3,900	108	1,080
.10	900	9,000	260	3,600	200	1,000
.20	1,720	8,600	690	3,450	390	950
.41	2,500	6,200	1,050	2,560	840	840
1.00	4,000	4,000	1,920	1,920	1,320	660
2.00	6,000	3,000	2,850	1,435	2,000	500
4.00	8,400	2,100	4,100	1,020	3,500	350
10.00	13,000	1,300	6,300	630		

[1] Air at 75° F. through a 1/8-in.-thick porous stainless steel sheet.

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[14] Rowley, F. B., and Jordon, R. C., Air Filter Performance as Affected by Low Rate of Dust Feed, Various Types of Carbon, and Dust Particle Size and Density: Trans. ASHVE, vol. 45, 1939, 339 pp.

TABLE 5. - Effect of temperature on pressure drop[1]

Dust fed, gr./sq. ft.	Δp, in. Hg		
	75° F.	1,000° F.	Ratio
0	0.051	0.22	6.4
100	.058	.30	5.2
1,000	.14	1.0	7.0
10,000	3.7	11.0	2.9

[1] Grade D filter; concentration of dust in feed gas, 1,000 gr./100 std. c.f.; specific gas flow, (1,000 std. c.f./hr.)/sq. ft. cross-sectional area of filter.

According to the impingement hypothesis, the increased kinetic energy of the dust particles at high temperature should tend to increase the percentage of dust removed because of greater statistical chance for impingement, but the percentage removed actually decreased (table 6). This decrease was caused by the larger gas volume, consequently the higher velocity of the gas, and by the higher viscosity of the gas. This agrees with First's results.[15]

TABLE 6. - Effect of temperature on percentage of dust removed[1]

Dust fed, gr./sq. ft.	Dust removed, percent	
	75° F.	1,000. F.
100	94.3	86.3
1,000	95.4	88.3
10,000	99.5	98.3

[1] Grade D filter; concentration of dust in feed gas, 1,000 gr./100 std. c.f.; gas flow, (1,000 std. c.f./hr.)/sq. ft. cross-sectional area of filter.

REGENERATION BY BLOWBACK

The principal advantage of porous stainless steel filters over fiber filters is that dust can be removed more easily by reversing the flow of gas. In the case of the stainless steel filter, a blast of a few seconds was as effective as a longer blast at the same rate. Experiments with glass tubes showed that long low-velocity blasts tend to agglomerate the dust, making it difficult to remove. Consequently, the blast should be applied suddenly rather than increased gradually. In practical applications this blast could be obtained from a storage tank, thereby removing the need for a high-capacity blower. A second blast did not decrease the pressure drop further and, except in one instance, did not remove any more dust from the filter.

Figure 10 shows the effectiveness of blowback flow for reducing the pressure drop built up during the dust-removal run. The concentration of dust in the gas during the filter run had little effect on this, but the runs at different concentrations can be distinguished in figure 10 by the different symbols used for the points. Percentagewise, the final pressure

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[15] Work cited in footnote 13.

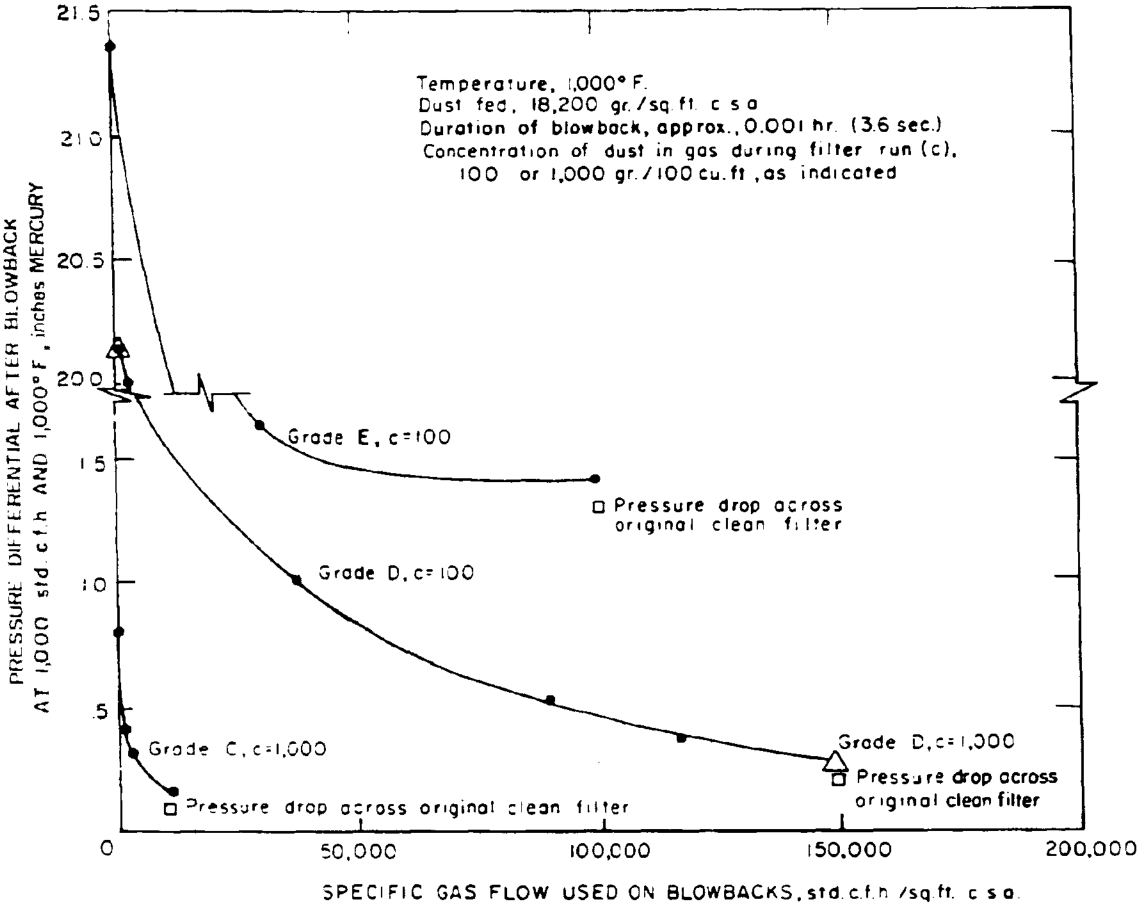

FIGURE 10. - Regeneration of Filters by Blowing Back to Reduce Pressure Drop.

drop after blowback was very close to the original pressure drop of the clean filter, except for the grade C filter. A flow of 4,000 std. c.f.h./sq. of cross-sectional area was enough to reduce by 90 percent the increase in pressure drop which occurred during the run. A flow of 100,000 std. c.f.h./sq. ft. of cross-sectional area reduced the pressure drop by 99 pct. Thus, the blowback effectively reduced the pressure drop across the filter.

Figure 11 shows the effectiveness of blowback for removing dust from the filter. In all instances a flow of 100,000 cu.ft./hr./sq. ft. of cross-sectional area accomplished virtually maximum removal of dust, while 50,000 was quite satisfactory except for the tightest filter, grade E. As might be expected, blowback effectiveness differed very little, whether the concentration of dust in the feed gas was 100 or 1,000 gr./100 cu. ft. The quantity of dust remaining in the filter after blowback was virtually the same for all grades--about 1,500 gr./sq. ft. of cross-sectional area. This quantity was equal to about 8 pct. of the filter volume, based on the absolute

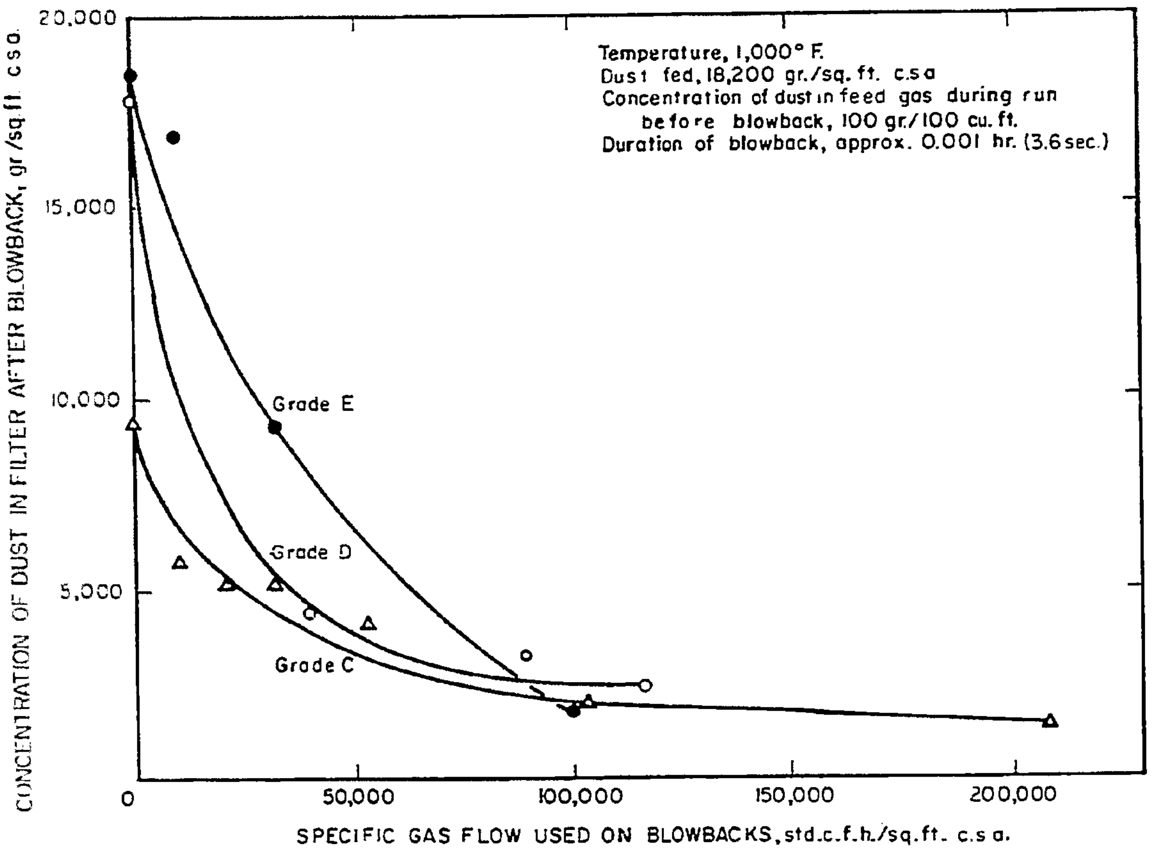

FIGURE 11. - Effect of Gas Flow During Blowback on the Removal of Dust From Different Grades of Porous Stainless Steel Filters.

density of the ash, or 25 pct. based on its bulk density. Practically, this quantity appeared to be irreducible by the blowback method.

Temperature had little effect on the ability of blowbacks to decrease pressure drop and remove dust from the filter (table 7). In this table the concentration of dust in the feed gas during the filtration run was different in the two instances but, as was shown previously, this had no effect on the results.

To test the practicability of repeated filtration and regeneration, such as would be desirable in actual practice, the apparatus was operated through a second filtration-blowback cycle. Results showed that pressure drop during a second filtration, at all dust loadings on the filter, was only slightly higher than during the first, and that a second blowback reduced the pressure drop to about the value for the clean filter (table 8). Pall showed that repeated blowbacks gradually increased the pressure drop, an increase that could be lessened by low gas flows during blowbacks.[16]

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[16] Pall, D. B., Filtration of Fluid Catalyst Lines from Effluent Gases: Ind. Eng. Chem., vol. 45, 1953, pp. 1197-1202.

TABLE 7. - Effect of temperature on the regeneration of grade D filters by blowing back[1]

	Δp across filter, in. Hg		Concentration of dust in filter, gr./sq. ft.	
	75° F.	1,000° F.	75° F.	1,000° F.
Original clean filter................	0.05	0.23	-	-
After filter run.....................	7.00	20.20	18,000	18,000
After blowback:				
34,000...............standard cubic feet per hour per square foot	.12	1.00	4,046	4,300
88,000....................do......	.08	.51	1,617	3,289
139,000...................do......	.07	.37	1,417	2,431

[1] Concentration of dust in feed gas: 75° F. run, 1,000 gr./100 cu. ft.; 1,000° F. run, 100 gr./100 cu. ft.; gas flow during filter run, 1,000 std. c.f.h./sq. ft.

TABLE 8. - Pressure differentials for first and second filtrations and blowbacks

Dust fed, gr./sq. ft.	Filtration, Δp, in. Hg	
	First	Second
0................................	0.096	0.110
200..............................	.110	.126
1,800............................	.167	.160
18,200...........................	.440	.634
After blowback...................	.110	.124

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