Bulletin 498
BUREAU OF MINES

LURGI PROCESS

USE FOR COMPLETE GASIFICATION OF COALS WITH STEAM AND OXYGEN UNDER PRESSURE

By J. Cooperman, J. D. Davis, W. Seymour, and W. L. Ruckes

UNITED STATES GOVERNMENT PRINTING OFFICE, WASHINGTON : 1951

UNITED STATES DEPARTMENT OF THE INTERIOR
Oscar L. Chapman, Secretary

BUREAU OF MINES
James Boyd, Director

For sale by the Superintendent of Documents, U. S. Government Printing Office
Washington 25, D. C.—Price 45 cents

CONTENTS

ILLUSTRATIONS

TABLES

LURGI PROCESS[1]

USE FOR COMPLETE GASIFICATION OF COALS WITH STEAM AND OXYGEN UNDER PRESSURE

By

J. Cooperman,[2] J. D. Davis,[3] W. Seymour,[3] and W. L. Ruckes[3]

Summary and Conclusions

THE INVESTIGATION was carried out in three stages, depending on the objectives of the work and the kind of equipment used. The first stage involved testing of 11 representative Alabama coals, the source of which is given in table 1. The objective was to find whether these coals could be gasified raw by the Lurgi method, and the equipment used was a 4-inch batch generator (to be described later). This generator proved too small and difficult to control to provide adequate amounts of products under continuous operating conditions for testing. For the second stage, a 6-inch batch generator was used, which was provided with facilities for superheating the steam-oxygen mixture and for synthesizing additional methane from the residual carbon monoxide and hydrogen in the primary Lurgi gas. Selected Alabama coals, Pittsburgh-bed coal from West Virginia, "Disco," and anthracite were used. For the third stage of the work, a continuous generator of 1 square foot grate area, following closely the design of the industrial generators operated in Germany, was provided. This generator was designed by O. Hubmann, engineer for the Lurgi company, and was expected to give results closely simulating industrial-scale apparatus. The purpose of this equipment was to test prepared materials shown to operate smoothly in the 6-inch generator under conditions approximating as nearly as possible those prevailing in industrial practice.

Work of the first stage showed that Alabama coals could not be used in the Lurgi process without first destroying the coking power. Coking in the generator restricted the flow of gases through the fuel bed and caused excessive channeling. Clinkering of the ash also presented a problem because all the fuels used (except anthracite) had low ash-fusion temperatures. The problem of caking was solved in the second stage by preliminary low-temperature carbonization of the coals, using a modification of the "Disco" process. Preoxidation was tried without success. The problem of clinkering was solved in the third stage by suitable modification of the automatic grate and control of the reaction temperature within appropriate limits. The following conclusions are drawn from results of the work:

1. Caking coals cannot be gasified by the Lurgi process. Coals that are weakly coking or noncoking may become coking under the operating conditions of this process and thus become useless without prior treatment.
2. Noncaking fuels, such as anthracite and low- and high-temperature cokes, can be completely gasified. Low-temperature coke or char can be made from bituminous coals in various ways. For the gasification tests described, it was made in batches in a rotating retort.
3. Although it was possible to gasify samples having a wide range of sizes, the optimum size range in the experimental generator was ⅛- by ¾-inch.
4. With the proper fuel size (⅛- by ¾-inch) a capacity as high as 9,000 cubic feet per hour per square foot of gasification area was obtained. However, when operating at this rate, the capacity of the steam boiler was exceeded, and the test had to be discontinued. A complete test was made at a capacity of 7,250 cubic feet per hour per square foot.
5. The heating value of the CO_2-free gas

[1] Work on manuscript completed June 1950.
[2] Chemist, Central Experiment Station, Bureau of Mines, Pittsburgh, Pa.
[3] Chemical engineer, Central Experiment Station, Bureau of Mines, Pittsburgh, Pa.

made from bituminous char was about 410 B. t. u. per cubic foot, which dropped slightly at high capacities.

6. The nickel methanization catalyst developed by the Gas Research Board in England was tested for activity and was found to make, from the Lurgi gas, a synthesis gas of about 950 B. t. u. per cubic foot after the CO_2 was scrubbed out. No attempt was made to determine the life of the catalyst.

7. The soda–iron oxide catalyst used to convert and remove the organic sulfur in the gas before synthesis was found to be a rather slow-acting material and required 10 to 15 times the volume of the nickel synthesis catalyst to purify a unit volume of gas.

INTRODUCTION

The ever-increasing demand for gas, natural or manufactured, for domestic and industrial use, plus rising costs in gathering and distributing natural gas over long distances and the prospect of eventual exhaustion of this fuel, has prompted many gas companies to turn serious attention to the possibility of making gas more cheaply from coal, which, relatively speaking, is in abundant supply. The intermittent water-gas process of making gas from coal is inherently expensive and has been unable to compete with natural gas, because, among other reasons, it generally requires expensive coke, oil, or oil products for enrichment. Of all types of commercial units in operation for making gas from coal, the one developed by the Lurgi company in Germany seems most promising from a competitive standpoint, since this process operates under pressure and the methane formed results in a manufactured gas of a higher heating value than that make in nonpressure processes. However, no information was available on whether this process would function satisfactorily with coals in the United States. Accordingly, the Southern Natural Gas Co. entered into an agreement with the Bureau of Mines to test some coals in small pressure units, using oxygen and steam as the gasifying agents. The United States Department of the Interior, Bureau of Mines, is not only interested in coal gasification for the manufacture of gaseous fuels but also in the raw gas as a possible source of materials for the Synthetic Liquid Fuels program.

ACKNOWLEDGMENTS

Thanks are due for the assistance given by C. R. Breck and Dr. C. Hochschwender of the Southern Natural Gas Co.; to the personnel connected with this project, consisting of W. E. Hake, W. F. Dugan, A. A. Wroblewski, W. C. McCall, R. D. Van Gilder, and F. Fischler; to H. M. Cooper, in charge of the coal-analysis laboratory, for the analyses of the coal samples; to D. E. Wolfson, in charge of the tar-analysis laboratory, for the analyses of the tar samples; to R. E. Brewer, in charge of the coal-oxidation laboratory, for the use of the oxidation equipment and for determining the plastic properties of the coal samples; and to M. H. Cizmarik and V. A. Hoysan, stenographers, for their assistance in handling the manuscript. Indebtedness is also acknowledged to L. L. Newman of the Bureau's staff for valuable suggestions during the progress of the work and for final review of the paper.

DISCUSSION

On November 1, 1945, an agreement was reached between the Bureau of Mines and the Southern Natural Gas Co. for investigating the possibilities of gasifying Alabama coals by the Lurgi process. This process, developed by the Lurgi company in Germany, completely gasifies carbonaceous materials under pressure, using oxygen and steam as the gasifying agents. In Germany, the commercial generator is usually operated at about 20 atmospheres pressure, although tests with pressures as high as 30 atmospheres have been made. Figure 2 shows a diagram of a commercial Lurgi high-pressure generator operating in Germany. Besides the obvious advantage of using oxygen instead of air, the Lurgi process working under pressure has the additional advantage that the reactions for the formation of methane are favored and, as a result, it produces a washed gas of about 450 B. t. u. per cubic foot. The theoretical effect of pressure on the gas composition is shown in figure 1. Reports from Germany on the Lurgi process indicate satisfactory operation with dried brown coals, lignites, and noncoking bituminous coals. However, the term "noncoking" was ill-defined in the German report, and it was believed that coals with poor coking properties might work in the Lurgi process. For theoretical reviews and actual operating results in Germany, the following references may be consulted.[4] [5] [6] [7] [8] [9]

[4] British Intelligence Objectives, Lurgi High-Pressure Gasification: BIOS Final Rept. 521, item 30, 56 pp.
[5] Newman, L. L., Oxygen in the Production of Hydrogen or Synthesis Gas: Ind. Eng. Chem., vol. 40, 1948, pp. 559–582.
[6] Odell, W. W., Gasification of Solid Fuels in Germany by the Lurgi, Winkler, and Leuna Slagging-Type Gas-Producer Processes: Bureau of Mines Inf. Circ. 7415, 46 pp.
[7] Weir, H. M., High-Pressure Gasification of Coal in Germany: Ind. Eng. Chem., vol. 29, 1947, pp. 48–54.
[8] Wagman, D. D., Kilpatrick, J. E., Taylor, W. J., Pitzer, K. S., and Rossini, F. D., Heats, Free Energies, and Equilibrium Constants of Some Reactions Involving O_2, H_2, H_2O, C, CO, CO_2, and CH_4: National Bureau of Standards, Journal of Research, Research Paper RP-1634, vol. 34, February 1945, pp. 143–161.
[9] Danulat, F., Mitt. Metallgas., No. 13, 1938, pp. 14–22.

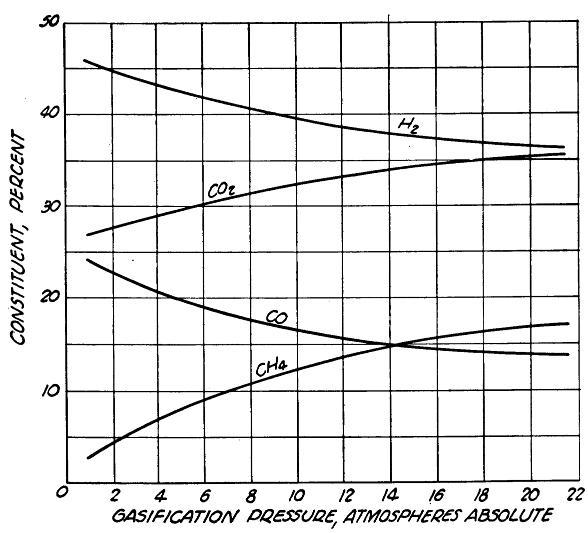

FIGURE 1.—COMPOSITION OF CRUDE GAS AS A FUNCTION OF PRESSURE (THEORETICAL).

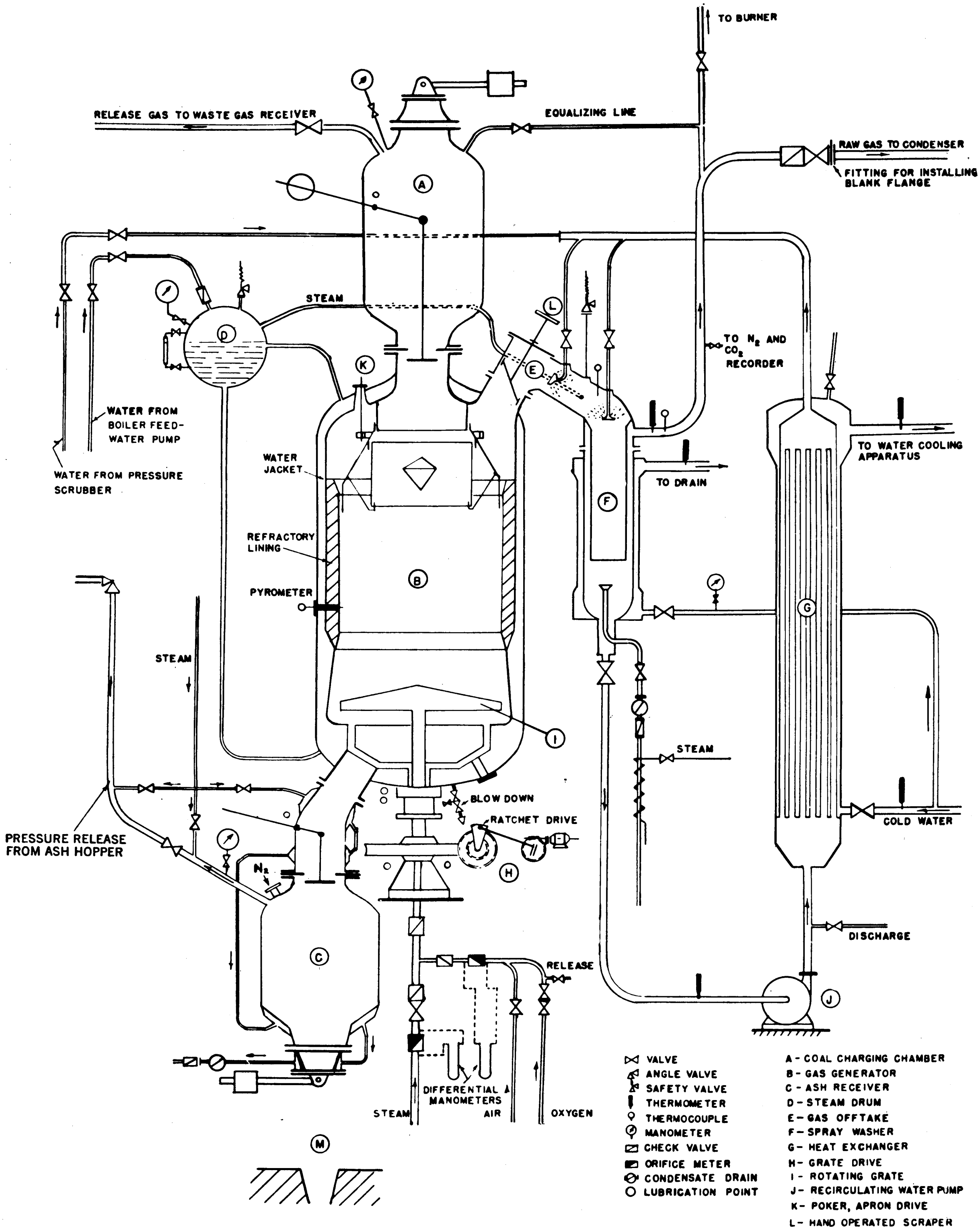

FIGURE 2.—DIAGRAM OF A COMMERCIAL LURGI HIGH-PRESSURE GENERATOR OPERATING IN GERMANY.

COALS TESTED

Samples were taken from various mines in the Alabama coal fields and sent to the Central Experiment Station of the Bureau of Mines for test. The samples are described in table 1. On receipt the coals were prepared for testing by crushing and screening to minus ½-inch and over 9-mesh (Tyler standard sieves). The portion passing through the 9-mesh sieve was discarded. Complete analyses were made of each sample; the results are tabulated in table 2. Plastic properties and agglutinating values are given in table 3. For the first series of tests, no attempts were made to lessen or destroy the coking properties of the coals.

TABLE 1.—*Coals received from Alabama at the Pittsburgh Station for Lurgi tests*

Coal No.	Description				Remarks
	Mine	Seam	County	Company	
264	Snider	Carter	Tuscaloosa	Snider Coal	These samples represent the Brookwood group. In general, both the mines and the production of this district are small. However, the coal reserves are not well-explored and may be large, and it was believed that the gas-making properties of these coals should be determined.
265	Abston	Milldale	do	Abston Coal	
266	do	Brookwood	do	do	
267	Warrior River	Mary Lee	Walker	Brookside-Pratt	These samples represent the free-burning coals found in Walker and Marion Counties. They are in the Warrior field, and existing data indicate that these coals should be suitable for use in gas-making under the proposed conditions. Since the Black Creek seam generally runs about 2 feet in thickness, it is sometimes mined by stripping operations.
268	New River	Black Creek	Marion	do	
269	Aldridge Shaft	Mary Lee	Walker	Stith Coal	
270	Deepwater	Black Creek	do	Brookside-Pratt	
271	Piper No. 2	Thompson	Bibb	Little Cahaba	These samples represent the coals found in the Cahaba field. They are not known as strongly coking coals.
272	Dogwood	Montevallo	Shelby	Little Gem	
273	Acmar	Mammoth	St. Clair	Alabama Fuel & Iron.	
275	Virginia	American	Jefferson	Republic Steel	This sample is a good coking coal that swells during carbonization. It was sampled and tested so that a comparison could be obtained between the other coals listed and this one of known good coking properties.

TABLE 2.—*Analyses of coals tested, as-received basis* [1]

Coal No.	Seam and mine	Proximate, percent				Ultimate, percent					Ash fusion, °F.			Heating value, B. t. u. per pound
		Moisture	Volatile matter	Fixed carbon	Ash	Hydrogen	Carbon	Nitrogen	Oxygen	Sulfur	Initial deformation	Softening	Fluid	
264	Carter seam, Snider mine	2.9	35.1	55.2	6.8	5.2	76.2	1.3	7.9	2.6	2,050	2,100	2,470	13,650
265	Milldale seam, Abston mine	1.8	34.6	56.0	7.6	5.2	77.9	1.5	6.9	.9	2,470	2,530	2,690	13,900
266	Brookwood seam, Abston mine	1.9	32.1	53.1	12.9	4.9	72.3	1.5	7.3	1.1	2,820	2,910+	--------	12,880
267	Mary Lee seam, Warrior River mine	3.5	34.1	48.4	14.0	5.0	67.9	1.6	10.2	1.3	2,570	2,660	2,760	12,160
268	Black Creek seam, New River mine	4.2	39.8	53.3	2.7	5.7	77.2	1.8	11.7	.9	2,230	2,320	2,620	13,800
269	Mary Lee seam, Aldridge Shaft mine	2.5	31.6	51.5	14.4	4.9	70.0	1.6	8.4	.7	2,850	2,910+	--------	12,400
270	Black Creek seam, Deepwater mine	3.8	40.0	51.9	4.3	5.7	76.5	1.7	10.5	1.3	2,220	2,310	2,610	13,720
271	Thompson seam, Piper No. 2 mine	2.8	39.2	53.5	4.5	5.4	78.1	1.2	10.3	.5	2,310	2,420	2,540	13,830
272	Montevallo seam, Dogwood Nos. 1 and 3	2.5	38.4	53.4	5.7	5.4	78.1	1.1	9.2	.5	2,310	2,440	2,540	13,850
273	Mammoth Nos. 2 and 3, Acmar mine	3.2	32.1	52.3	12.4	5.1	72.0	1.5	8.4	.6	2,680	2,740	2,810	12,850
275	American seam, Virginia mine	3.4	21.7	70.6	4.3	4.9	82.9	1.4	6.0	.5	2,680	2,760	2,910+	14,510

[1] Analyses made under supervision of H. M. Cooper, chemist, Central Experiment Station, Bureau of Mines, Pittsburgh, Pa.

TABLE 3.—*Plastic and agglutinating properties of coal*

Coal No.	Davis plastometer, resistance develops, °C.	Maximum fluidity, Davis plastometer, minimum resistance		Solidification, Davis plastometer, maximum resistance		Davis plastometer resistance ends, °C.	Agglutinating values (silicon carbide: coal ratio, 15:1)
		°C.	Lb.-in.	°C.	Lb.-in.		
264	406	445–454	0.3	467	10.7	521	6.4
265	404	424–463	.3	477	6.4	491	6.5
266	404	421–458	.3	476	17.9	499	7.5
267	419	430	.7	451	20.0	497	5.3
268	----------	(1)	----------	540	.7	564	3.2
269	418	437–448	.4	469	22.4	510	6.0
270	421	436–446	.2	460	7.2	496	4.9
271	416	435	.7	455	7.9	477	5.3
272	421	(2)	----------	423	2.1	457	6.6
273	412	428–457	.4	465	6.2	477	7.9
275	425	457–480	.2	499	36.5	554	8.3

[1] Very slight fusion.
[2] Rising curve.

DESCRIPTION OF AND RESULTS WITH 4-INCH BATCH GENERATOR

An experimental generator was designed and assembled. The generator proper consisted of an inner 4-inch stainless-steel and an outer 6-inch steel pipe, so constructed that gasification of the coal took place in the inner pipe while the pressure in the generator was held by the relatively cool outer pipe. (See fig. 3.) A revolving grate, an inlet for the oxygen-steam mixture, and an ash hopper, with valve and cap arrangement to discharge ash under pressure, were incorporated at the bottom of the generator. A gas outlet, a valve, a charging hopper, and a cap were constructed at the top to permit charging when operating under pressure. Eight thermocouples were impinged against the inner pipe at various distances along its length. The generator with the various meters and apparatus was completely assembled and ready for operation on March 1, 1946.

Table 4 summarizes the test data for 31 tests in the 4-inch generator. The first test, made with Disco—a low-temperature coke manufactured by the Disco Co.—had to be discontinued soon after the start because of trouble with the ash-removal mechanism. (The analysis of Disco is given in table 11.) The mechanism was repaired and a second test started. This test, at atmospheric pressure, was successful; the data obtained are recorded in table 4. Interesting observations were that the combustion

TABLE 4.—*Results with 4-inch-internal-diameter generator*

Test No.	Coal No.	Treatment	Fuel, lb.	Oxygen, cu. ft.[1]	Steam, lb.	Gas produced, cu. ft.[1]	Analysis, percent			Remarks
							CO_2	H_2	CO	
1	Disco 307 [2]	None								Discontinued due to trouble with ash-removal mechanism.
2	do	do	17.0	139	13¾	586	10.9	36.5	43.8	
3	do	do	17.5	178	15¾	606	No analysis			Line clogged.
4	268	do		133	9	125	do			40 percent O_2 in effluent gas.
5	268	do	8.0	175	12⅛	180	do			Charge coked and stuck.
6	268	do	19.3	135	8	379	11.8	31.8	49.2	Charge poked.
7	264	do	18.5	170	12⅝	518	11.2	28.0	49.0	Do.
8	273	do								Discontinued when inner liner burned through.
9	273	do	10.0	113	10¾	297	24.4	28.8	42.0	Outer 6-inch tube removed.
10	265	do	6.3	76	6½	178	30.6	27.1	31.2	Charge poked.
11	266	do	3.3	35	3⅝	72	36.5	21.6	32.7	Do.
12	267	do	9.3	97	6½	265	No sample			
13	269	do	8.5	94	7¼	250	25.2	28.3	31.0	Do.
14	270	do	11.0	114	14¼	322	24.8	32.0	33.9	Do.
15	271	do	9.0	97	5½	244	26.4	28.6	28.0	Do.
16	272	do	9.0	114	5¾	278	No sample			
17	275	do	4.8	90	5¼	180	20.5	33.2	17.0	Do.
18	Barley anthracite 318	do	9.8	114	7⅛	310	19.6	34.4	34.9	Gasified well.
19	268–1ox [3]	24 hr. under O_2 at 100° C.	7.4	81	5½	215	27.8	20.8	27.0	Charge poked.
20	268–2ox [3]	48 hr. under O_2 at 100° C.	7.3	80	5	237	24.8	25.5	36.7	Do.
21	270–1ox [3]	24 hr. under O_2 at 100° C	8.3	76	7¼	227	23.6	27.8	31.5	Do.
22	270–2ox [3]	84 hr. under O_2 at 100° C	8.5	79	7¾	256	23.1	28.9	36.6	Do.
23	268–3ox [3]	101 hr. under O_2 at 100° C	8.3	89	3⅜	223	27.1	20.2	44.6	Poked. Gas channeled.
24	184 [4]	None	8.0	90	6⅝	165	40.8	19.1	33.2	Gas channeled up the sides of the tube.
25	268–4ox [3]	141 hr. under O_2 at 100° C	7.5	77	5⅛	246	21.0	24.6	37.2	Sample 0.25 by 0 inch poked.
26	268–4ox [3]	do	4.0	115	7¾	166	55.8	16.2	10.9	Generator redesigned to prevent channeling. Could not poke.
27	270–4x [5]	Low-temperature char [6]	13.0	132	3⅞	435	8.3	33.5	48.7	No poking required.
28	270–5x [5]	do								Discontinued due to burn-out.
29	270–6x [5]	do	9.0	94	8½	385	14.9	33.7	41.2	No poking required.
30	268–7x plus [5] 25 percent raw 238	do	8.4	102	7	312	19.9	24.9	38.4	Some poking required.
31	270–7x [5]	do	9.0	75	7¾	405	No analysis			Sample ½ by 0 inch gasified well.

[1] 30 inch Hg and 60° F. dry.
[2] See table 11 for analysis.
[3] Coal had been oxidized, for various times indicated.
[4] Anthracite fines; see table 11 for analysis and table 12 for screen size.
[5] Coal had been carbonized.
[6] See table 5, tests E and F.

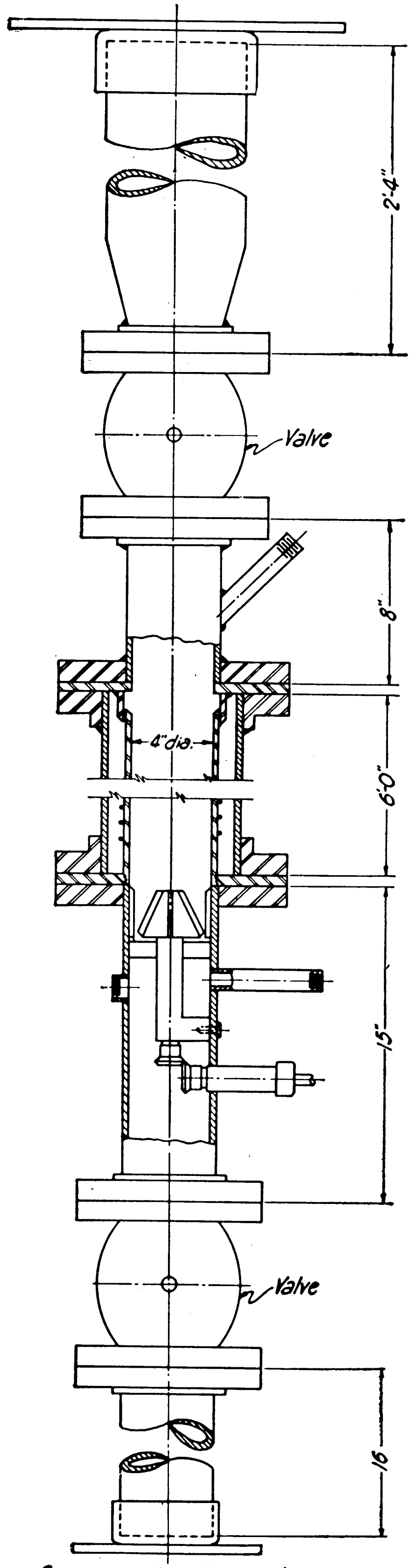

FIGURE 3.—4-INCH BATCH GENERATOR.

took place in a relatively narrow zone and that the temperature dropped rapidly in the upper part of the generator.

After various changes and repairs, the first test (test 4) with Alabama coal was made on March 15, 1946. Coal 268, from the Black Creek seam, was selected for this test because the analysis indicated that, of all the coals received, this one probably would be the most satisfactory. In this test, the pressure at the bottom of the generator built up quickly to about 15 pounds per square inch; the coal did not feed down through the generator, and the CO_2 contents of the gas remained high. During the latter part of the run the outlet gas contained an excessive amount of oxygen, indicating that it was bypassing the inner pipe. Examination of the generator after the shut-down showed an empty space 14 inches high at the bottom of the pipe. Above this was 16 inches of solid coke followed by coal in which some tar had condensed. Various changes in the apparatus and in operating procedure were made in all the following tests using coking coals, but the only successful runs were those in which the charge was poked with a poker incorporated in the generator. The poking broke up coke formation, and only then did gasification proceed satisfactorily. No attempt was made to operate under pressure, since it would have then been impossible to poke the charge by hand. With anthracite, no poking was necessary, and no trouble was experienced in operation. In test 18, with Barley anthracite, the gasification proceeded in a manner similar to the tests made with Disco. With the anthracite fines in test 24, there were indications of channeling.

After test 8, when the inner pipe had burned through, the generator was rebuilt without the outside 6-inch pipe, since it was apparent that no pressure tests would be possible with coking coals. Without the outside pipe, it was easier to follow the combustion zone as well as to observe channeling of the gases by the hot spots formed on the 4-inch pipe. Although it was now possible to operate the generator without the danger of burn-outs, the greater heat loss resulted in a gas with a higher CO_2 and lower CO content. This effect is shown in table 4 by comparing the gas analyses in the first seven tests with those in the following tests. The small generator with its relatively large heat losses, plus the absence of a superheater for the steam and oxygen, made a gas with an unfavorable H_2 : CO ratio for the synthesis of methane.

As a result of the above series of tests, the conclusion was drawn that coals with any coking tendencies could not be used in the Lurgi process.

DESTRUCTION OF COKING PROPERTIES

The next step in the research program was to investigate methods of destroying the coking properties of coals. Two general methods (oxidation and low-temperature carbonization) were tried.

OXIDATION OF COALS

The Coal Carbonization Section of the Central Experiment Station, in the course of its investigation of coking coals, has developed rotating retorts for studying the effect of air and oxygen at temperatures up to 100° C. on the coking properties of coals.[10] Since coals 268 and 270 had the poorest coking properties of the series of Alabama coals, they were oxidized and used in the gasification tests recorded in table 4. Coal 268 (tests 19 to 26) was finally oxidized with oxygen at 100° C. for as long as 141 hours without losing all of its coking properties. Tests 25 and 26 (table 4) indicated that oxidizing a coal which is just coarse enough to prevent channeling in the generator did not completely destroy all of its coking properties. When the furnace bottom was changed to give the incoming oxygen and steam a better chance for distribution before reaction with the oxidized coal, poking was still required for gasification. The coke formed was porous but held together enough to prevent the charge from moving down.

The gasification tests with the above-mentioned coals oxidized at 100° C. (table 4) demonstrated that more drastic treatment was required to produce a product that would gasify satisfactorily in the generator. It was, therefore, decided to try oxidation at a higher temperature. For this purpose a Franz Fischer rotary retort, which had been used for low-temperature carbonization by the Coal Carbonization Section, was available. This apparatus consists of: (1) A steel-drum retort, 12 inches internal diameter and 43 inches long, with removable heads and with swivel joints for air inlet and gas outlet; (2) a trunnion with motor drive for rotating the drum; (3) a gas burner under the drum for heating; and (4) a shield over the drum to provide some insulation. (See fig. 4.) The apparatus was assembled with a thermocouple installed through the inlet pipe, which dipped into the coal so as to measure actual coal temperatures. A metered air supply was connected to the inlet pipe, while the effluent gases were allowed to escape. Coals 268 and 270, which have been used in all previous oxidation tests, were again selected for testing at higher temperatures. As size is an important factor in oxidation, the coal was crushed to minus ¼-inch on 14-mesh (Tyler standard sieve). This size was chosen because it was probably the smallest size that would work satisfactorily in the generator. The minus 14-mesh was discarded.

The first sample of coal 268 (table 5, test A) was treated for a total time of 7.5 hours at a maximum temperature of 216° C. for 5 hours. Forty pounds of the sized coal was put in the drum, air added at an average rate of 100 cubic feet per hour, and the gas burner lit under the retort. A temperature of 200° C. was reached in 1 hour after the test was started and was maintained within the range of 196° to 216° C. A composite sample of the exit gas showed the following composition in volume percent: CO_2, 1; O_2, 17.9; CO, 0.4; CH_4, 0.12; and N_2, 80.6.

Instead of testing the oxidized samples for their gasification properties in the Lurgi generator, as has been done up to the present, a simpler method was devised to test the coking properties of the treated coals. A small retort was made from a 5-inch pipe about 8 inches long. The pipe was capped on both ends, and a ¾-inch pipe welded in one cap. The cap on the other end was removable. When a treated coal was to be tested, the small 5-inch retort was charged through the removable cap, the cap replaced, and the inverted retort heated in an electric furnace to 900° C. A flexible hose was attached to the ¾-inch pipe to remove gas and tar during the test.

After the coking test in the 5-inch retort had shown that oxidation of coal 268 at 216° C. (test A, table 5) did not destroy the coking property, a second test (B) was made at 305° C. This test again showed the sample sticking together. When the same result was obtained with coal 270 (test C), it was evident that still more drastic treatment was required. However, at a temperature of about 300° C. the heat produced by the reaction of air and coal was enough to slowly raise the temperature in the retort. Table 5 lists the samples tested and results obtained by coking in the test retort. Analyses of coals 268 and 270 after the various oxidation stages summarized in tables 4 and 5 are given in table 6.

[10] Schmidt, L. D., and Elder, J. L., Atmospheric Oxidation of Coal at Moderate Temperatures. Ind. Eng. Chem., vol. 32, February 1940, pp. 249–256.

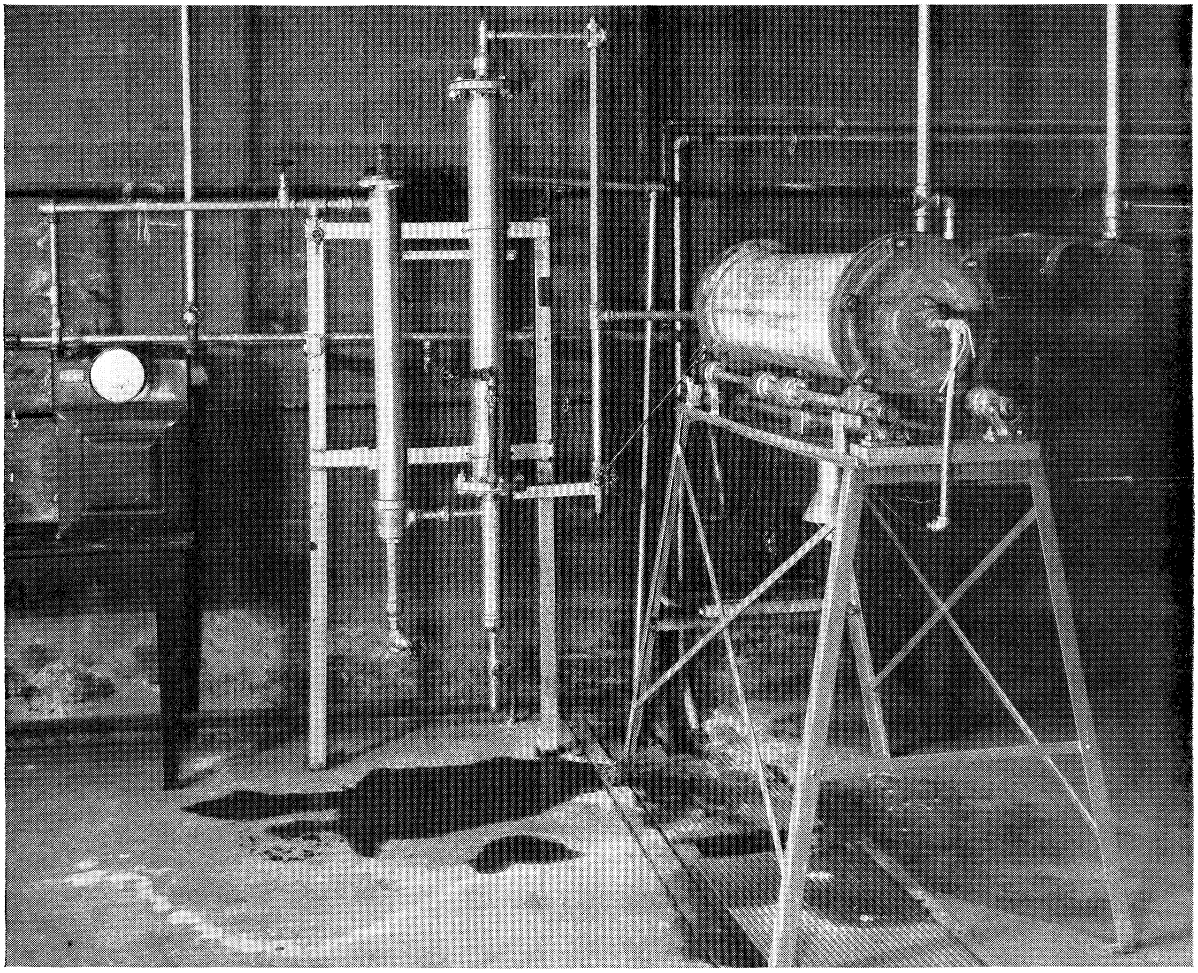

FIGURE 4.—FRANZ FISCHER ROTARY RETORT (LOW-TEMPERATURE CARBONIZER).

LOW-TEMPERATURE CARBONIZATION OF COALS

It was believed that heating the coal to a higher temperature in the presence of air would cause actual burning. The next step, therefore, was to heat the coal to a higher temperature in the absence of air. A 32-pound sample of coal 270 that had been oxidized for 101 hours at 100° C. (coal 270–3ox, table 5) was charged in the retort and heating without admission of air was started. After 55 minutes, the temperature reached 358° C., when considerable gas and some tar were evolved. The heating was continued, and the temperature gradually increased to 398° C., when the heating gas was turned off. After the retort had cooled to room temperature, it was opened and the residue examined. Partial coking had taken place. The product was small balls or distorted spheres, the largest being about ⅝ inch in diameter. There was no cohesion between balls. The weight of the recovered material was 27¾ pounds, which means that 4¼ pounds (13.3 percent) of volatile matter had been driven off. A coking test in the 5-inch retort at 900° C. indicated that the char had no coking properties, and it was concluded that this product should work in the Lurgi generator. Sample 270–4x was tried with the results as given in table 4, test 27. This test was started in the usual manner. The generator heated up very slowly, but the gas rate was exceptionally high. Analyses of grab samples showed low CO_2 content. No poking was required to keep the charge moving down. Examination of the generator after the test showed uniform combustion across the whole gasification area and no indication of the charge sticking to the generator wall.

A carbonization test made with unoxidized coal 270 (test E, table 5) gave virtually the same results as were obtained in test D using the oxidized coal. It was concluded that pre-

TABLE 5.—*High-temperature oxidation and low-temperature carbonization pretreatment of coals and small-retort tests of products*

Test No.	Coal No.	Size range [1]	Air passed over coal	Temp., °C.	Time at max. temp., hrs.	Total time, hrs.	Remarks on products
A	268	¼ in.–14-mesh	100 cu. ft. per hour.[2]	216	5. 0	7. 5	268–5ox coked in test retort.
B	268–5ox [3]	do	do	305	3. 5	5. 5	268–6ox coked in test retort.
C	270	do	do	305	4. 5	5. 5	270–3ox coked in test retort.
D	270–3ox	do	None	398	2. 5	3. 0	270–4x used in Lurgi test 27; noncoking.
E	270	do	do	403	1. 5	3. 5	270–5x used in Lurgi tests 28 and 29; noncoking.
F	270	Same as test E. Char used in Lurgi test 29.					270–6x.
G	275	¼ in.–14-mesh	None	402	2. 5	3. 0	Coked in retort test.
H	275	do	do	450	1. 5	3. 5	Noncoking; balls up to 2.5 in. formed.
I	294 [4]	⅛ in.–0	do	460	1. 0	2. 5	Sample melted and stuck to retort.
J	270	½ in.–0	do	424	1. 0	2. 5	Fines balled up; noncoking.
K	264	½ in.–9-mesh	do	440	. 5	3. 0	Coal heated too fast; large balls formed.
L	265+25 percent breeze.	¼ in.–14-mesh below ¼ in.	do	432	. 5	2. 5	Noncoking; coal without breeze melted and stuck to retort.
M	268	¼ in.–14-mesh	do	413	. 5	1. 5	Noncoking.
N	273	½ in.–9-mesh + 25 percent fines.	do	427	. 5	2. 0	Coal stuck to retort without fines; balls 3 in. by 0 formed.
O	272	do	do	401	1. 0	2. 0	Noncoking; balls 2 in. by 0 formed.

[1] Tyler standard screen-scale sieves.
[2] 30-inch Hg and 60° F. wet.
[3] Coal had been oxidized for various times stated.
[4] See table 11 for analysis.

TABLE 6.—*Analysis of oxidized coals (air-dried)*

Coal No.	Treatment	Analysis, percent						Remarks
		Hydrogen	Carbon	Nitrogen	Oxygen	Sulfur	Ash	
268–1ox	100° C. under O_2 for 24 hr	5. 6	77. 6	1. 7	11. 1	1. 1	2. 9	
268–2ox	100° C. under O_2 for 48 hr	5. 5	77. 3	1. 7	11. 5	1. 0	3. 0	
268–3ox	100° C. under O_2 for 101 hr	5. 5	76. 6	1. 7	12. 1	. 9	3. 2	
268–4ox	100° C. under O_2 for 141 hr	5. 5	76. 2	1. 8	12. 9	1. 0	2. 6	
268–5ox	216° C. under air for 7.5 hr	5. 2	76. 4	1. 8	13. 0	. 9	2. 7	(Offtake gas: CO_2, 4.2; O_2, 14.7; CO, 1.0; CH_4, 0.3; N_2, 79.8.)
268–6ox	305° C. under air for 5.5 hr	5. 1	77. 6	1. 7	12. 0	. 9	2. 7	
270–2ox	100° C. under O_2 for 84 hr	5. 4	76. 2	1. 7	11. 5	1. 4	3. 8	
270–3ox	305° C. under air for 5.5 hr	5. 1	77. 9	1. 7	9. 8	1. 3	4. 2	(Offtake gas: CO_2, 3.8; O_2, 12.2; CO, 1.3; CH_4, 0.3; N_2, 82.4.)

oxidation was not necessary in making low-temperature char. None of the remaining Alabama coals (table 5) were oxidized before they were carbonized. To measure the by-products of the low-temperature carbonization, a condenser, a Cottrell precipitator, and a gas meter were installed in a train after the retort. Tables 7 and 8 give the results of the low-temperature carbonization of the Alabama coals.

As a rule, each coal has different carbonization characteristics; variations in treatment are necessary to produce a char of the desired density and hardness for gasification. The variations consist of changes in the rate of heating, the time of heating, the maximum temperature required, and, in some cases, the addition of low-temperature fines. The main products recovered from the low-temperature distillation consist of char, tar, liquor, and gas. The quantity of each depends primarily upon the analysis of the coal. A high-volatile coal usually gives a higher yield of tar and gas than a low-volatile coal.

TABLE 7.—*Yields of products of low-temperature carbonization per ton of coal, as-carbonized basis*

Coal No.	Description and source	Dry tar per ton of coal, gallons	Gas per ton of coal, cu. ft.[1]	Char per ton of coal, lb.
267	Brookside-Pratt Mining Co., Warrior River mine	20. 4	940	1, 610
268	Brookside-Pratt Mining Co., New River mine	25. 0	940	1, 594
269	Stith Coal Co., Aldridge mine	20. 2	1, 140	1, 650
270	Brookside-Pratt Mining Co., Deepwater mine	23. 0	970	1, 588
271	Little Cahaba Coal Co., Piper No. 2 mine	25. 3	1, 292	1, 588
272	Little Gem Coal Co., Dogwood Nos. 1 and 3 mines	26. 0	1, 250	1, 580
264	Snider Coal Co., Snider mine			1, 600
265	Abston Coal Co., Abston mine, Milldale seam			1, 700
266	Abston Coal Co., Abston mine, Brookwood seam			1, 600
273	Alabama Fuel & Iron Co., Acmar Nos. 2 and 3 mines			1, 600
275	Republic Steel Corp., Virginia mine			1, 750

Coking tests on coals 264, 265, 266, 273, and 275 were made before the apparatus for collecting tar and measuring gas was available.

[1] B. t. u. of gas between 900 and 1,000 per cubic foot.

TABLE 8.—*Yields of products of low-temperature carbonization, as-carbonized basis*

Coal No.	Treatment	Char, percent	Tar (dry), percent	Liquor, percent	Gas,[1] percent	Char analysis, percent		
						Volatile	Fixed carbon	Ash
267	Carbonized at about 400° C	80. 5	8. 5	8. 1	3. 3	19. 1	63. 2	17. 7
268	do	79. 7	10. 4	7. 6	3. 3	21. 7	74. 0	4. 3
269	do	82. 5	8. 4	5. 5	4. 0	17. 4	65. 8	16. 8
270	do	79. 4	9. 6	6. 8	3. 5	23. 3	70. 9	5. 8
271	do	79. 4	10. 6	6. 1	4. 6	19. 7	74. 2	6. 1
272	do	79. 0	11. 0	5. 5	4. 4	20. 7	71. 7	7. 6

[1] Gas weight taken at 14 cubic feet per pound.

DESCRIPTION OF AND RESULTS WITH 6-INCH, HIGH-PRESSURE BATCH GENERATOR

Since a method has been found to destroy the coking properties of the Alabama coals, the question of testing these coals under pressure was again considered and it was decided to construct a second generator for the pressure tests. The present 4-inch generator had been so altered during the testing of the raw coals that it would have had to be rebuilt completely to operate under pressure. Since it was not feasible to operate such a small generator continuously, the second one (fig. 5) was designed as a batch unit and with a stainless-steel 6-inch instead of a 4-inch pipe for the gasification chamber. The outer shell was made from a piece of 12-inch pipe. Insulating material was placed between the two shells. The charging hopper was a 12-inch pipe, 3 feet long, connected to the generator proper by a 14-inch length of 6-inch pipe. As the coal gasified in the generator, the coal from the charging hopper fed down into the 6-inch gasification chamber. The gasification zone moved up slowly as the ash layer increased; the test was discontinued when it reached about halfway up the generator. The location of the hot zone was determined by a series of thermocouples extending through the outside 12-inch pipe to the outer wall of the 6-inch pipe. Temperatures were recorded by a recording potentiometer. The oxygen and steam were measured by flow meters, and the pressure in the generator was regulated by an automatic back-pressure-regulating valve. The gas leaving the generator was cooled in an indirect water-cooled condenser, expanded, and washed with a water spray before entering the displacement meter. The gas was sampled and then vented to the air. The supply of oxygen was obtained from a battery of five 200-cubic foot oxygen cylinders which were exhausted in two runs. An electrically heated high-pressure boiler was the source of the steam supply. Figure 6 is a photograph of the generator and accessory equipment.

The first test with the 6-inch batch generator was made on January 31, 1947, using Disco as fuel. This test (32) was more or less exploratory in nature to acquaint the personnel with the operation of the generator under pressure. The generator was operated at low capacity and under a maximum pressure of 150 pounds per square inch. Table 9 summarizes the test data obtained with this generator using eight different fuels. In test 33 the pressure was raised to 445 pounds per square inch. In the original design no provision was made for superheating

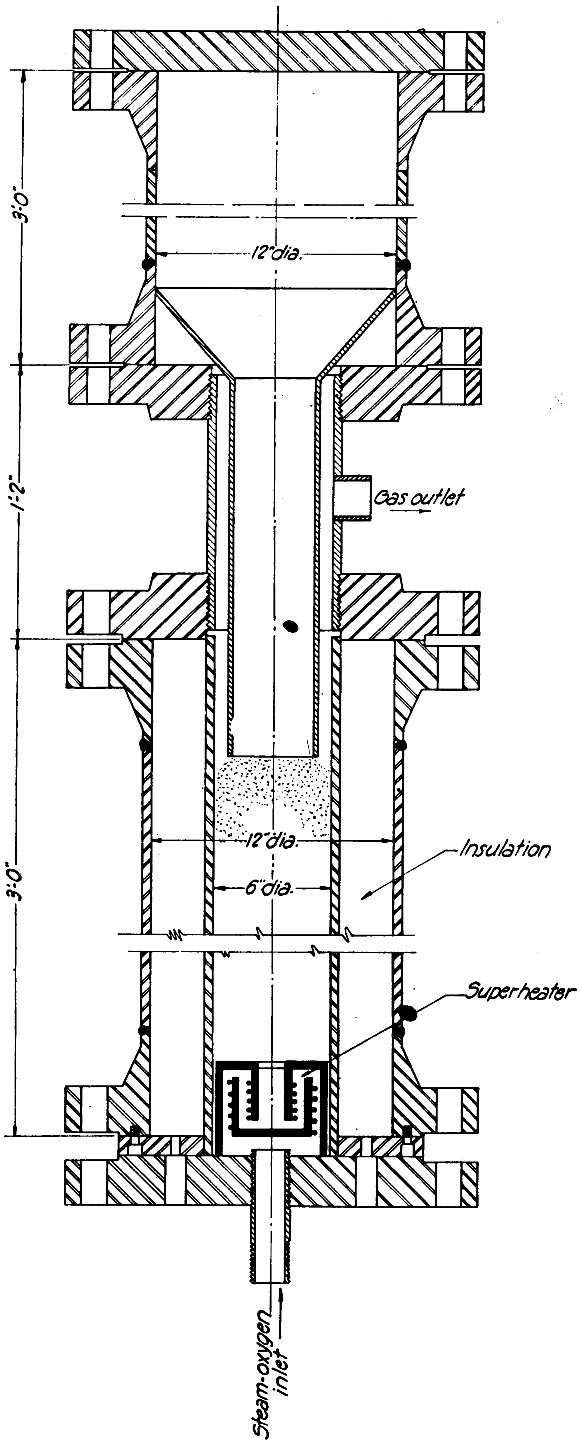

FIGURE 5.—6-INCH, HIGH-PRESSURE BATCH GENERATOR.

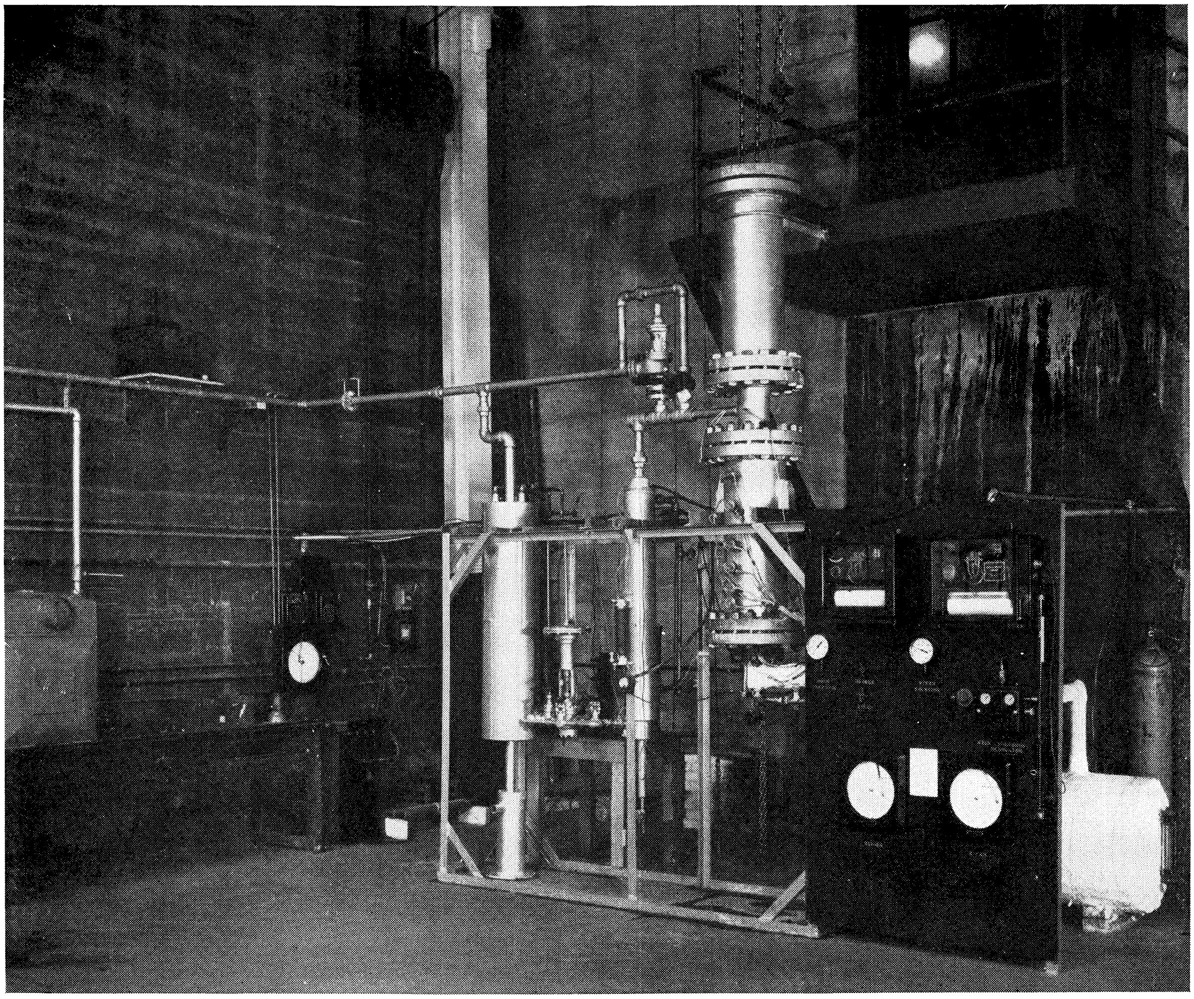

FIGURE 6.—6-INCH, HIGH-PRESSURE BATCH GENERATOR WITH ACCESSORY EQUIPMENT.

the oxygen and steam, and, as a result, the analysis of the effluent gas showed a low $H_2 : CO$ ratio. (See table 9.) To increase the H_2 content of the gas, an electrically heated superheater was installed in the lower portion of the 6-inch gasification pipe. Before each test a 3- to 4-inch layer of ½- by ¼-inch ceramic pieces was placed over the superheater to prevent the charge from burning at the superheater. Test 34 was made with the superheater operating and the generator at 300 pounds per square inch pressure, using Disco as fuel. Test 35 was made using semianthracite as the fuel. Semianthracite and anthracite (tests 46, 48, and 52) gave a gas with a somewhat lower methane content but gasified as well as char of comparable size. The difficulties experienced in gasifying anthracites were in tests with charge sizes mainly under ⅛-inch. The reactivity of coals of all grades approaches the same order of magnitude at about 2,000° F. At the lower temperature at which methane formation takes place, the chars showed much greater reactivity than did the anthracites. Extremely high reactivity of the chars was amply demonstrated qualitatively. Probably some of the methane found was contributed by the volatile of the chars, but gases yielded on recarbonization of chars is known to be very poor in methane. The duration of the tests made in this generator was about 3 hours.

The first pressure test with Alabama char was test 42. Char made from coal 268 was used. In this and the following tests with char, the gasification under pressure proceeded smoothly. However, in no case was a CO_2-free gas of much over 410 B. t. u. per cubic foot produced. Sodium carbonate was added to the charges in tests 44 and 46 with the hope of increasing the reactivity of the charge, but the CO_2-free gas produced was still about 410 B. t. u. In test 48 and 52, anthracites of

Buckwheat Nos. 4 and 5 sizes were used. The pressure was increased to 440 pounds per square inch in these tests to slow down the rate of gas flow through the fine charge and thus prevent channeling and formation of hot spots in the generator. The anthracite in Buckwheat No. 4 size (test 48) gasified smoothly, but with Buckwheat No. 5 (test 52) there were indications that the gases were channeling up through the charge.

TABLE 9—*Test data for 6-inch batch generator*

Test No.	33.	34.	35.	42.
Date	2-6-47.	2-19-47.	2-21-47.	4-9-47.
Time	11:02 a. m. to 2:00 p. m.	9:45 a. m. to 1:05 p. m.	10:45 a. m. to 12:30 p. m.	12:48 p. m. to 3:55 p. m.
Fuel, kind	Disco (307), ½ in. x 9 mesh.[1]	Disco (307), ½ in. x 9 mesh.[1]	Semianthracite (274),[2] ¼ in. x 14 mesh.[1]	Char (268).
Amount of fuel	43¾ lb.	42 lb.	29 lb.	35½ lb.
Oxygen	280 cu. ft.	280 cu. ft.	170 cu. ft.	228 cu. ft.
Steam	20 lb.	50 lb.	29 lb.	77 lb.
Gas produced	1,568 cu. ft.	Not measured.	Not measured.	1,450 cu. ft. N. T. P.

Test No.	33					34					35				42					
Time	11:55	12:10	12:40	1:10	1:40	10:54	11:50	12:23	12:35	12:55	11:30	11:45	12:00	12:30	1:30	2:00	2:30	3:00	3:30	3:55
Pressure, lb	------	100	200	300	445	300	300	300	300	300	310	300	300	300	300	300	300	300	300	300
CO_2 percent	9.1	15.4	16.3	16.1	15.4	17.5	19.7	23.6	26.8	25.1	12.5	14.2	15.6	14.6	27.2	25.5	26.8	29.9	28.7	29.7
Ill do	.9	.5	.8	.9	1.1	.4	.6	.4	.4	.4	1.6	1.4	1.5	1.0	.6	.8	1.0	1.2	1.0	.6
O_2 do	1.0	.2	.4	.0	.4	.7	.4	1.1	.4	.4	1.0	.2	.2	1.2	.5	.2	.2	.2	.4	.3
H_2 do	39.7	35.0	33.8	31.4	28.7	32.3	35.0	36.2	36.5	36.5	36.0	37.9	37.3	36.2	33.2	37.2	36.9	37.4	37.6	38.5
CO do	41.6	36.2	34.9	37.1	41.2	34.3	30.6	26.6	22.9	24.9	38.9	33.4	33.6	38.7	16.0	21.7	21.2	19.8	19.9	20.2
CH_4 do	1.4	7.2	7.6	9.1	7.2	10.6	8.7	8.3	7.4	7.5	4.7	6.2	6.0	3.0	14.3	9.4	8.4	8.8	6.0	8.3
C_2H_6 do	1.5	.5	.8	.0	.7	.0	.0	.4	.4	.6	1.2	.8	.6	.8	1.9	1.1	1.0	.2	2.0	1.3
N_2 do	4.8	5.0	5.4	5.4	5.3	4.2	5.0	3.4	5.2	4.6	4.1	5.9	5.2	4.5	6.3	4.1	4.5	2.5	4.4	1.1

Test No.	44.	46.	48.	52.
Date	4-25-47.	5-22-47.	6-10-47.	7-16-47.
Time	9:25 a. m. to 12 noon.	12:40 p. m. to 2:50 p. m.	12:35 p. m. to 2:45 p. m.	1:15 to 3:15 p. m.
Fuel, kind	Activated L. T. coke (268).	Anthracite Barley (318), activated.[2]	Anthracite Buckwheat (319).[2]	Buckwheat (320).[2]
Amount of fuel	41¼ lb. (including 2 lb. Na_2CO_3).	36 lb., 2 lb. Na_2CO_3.	23 lb.	25 lb.
Oxygen	------	184 cu. ft.	192 cu. ft.	160 cu. ft.
Steam	78 lb.	60 lb.	55 lb.	53.9 lb.
Gas produced	1,688 cu. ft. N. T. P.	1,277 cu. ft. N. T. P.	879 cu. ft. N. T. P.	794 cu. ft. N. T. P.

Test No.	44					46				48				52			
Time	10:00	10:30	11:00	11:30	12:00	1:15	1:45	2:15	2:45	1:15	1:45	2:15	2:45	1:45	2:15	2:45	3:15
Pressure, lb	300	300	300	300	300	300	300	300	375	440	440	440	440	440	440	440	440
CO_2 percent	28.7	27.3	27.9	27.4	26.9	23.7	28.2	29.5	29.1	31.8	26.0	30.8	31.8	26.6	26.2	37.1	38.7
Ill do	.2	.2	.2	.2	.2	.8	.4	.2	.2	.3	.6	.2	.1	.8	.6	.4	.4
O_2 do	.2	.2	.2	.2	.2	.2	.2	.2	.4	.7	.4	.6	.3	.2	.4	.2	.2
H_2 do	42.6	42.6	43.3	43.1	43.0	44.5	43.7	42.9	41.5	37.0	36.9	33.9	32.6	38.5	36.1	30.2	30.0
CO do	15.5	16.7	16.7	16.8	17.4	21.6	18.9	19.0	20.8	22.6	24.5	23.7	25.3	21.7	21.9	19.1	18.8
CH_4 do	7.9	8.8	7.1	8.1	8.3	5.1	4.9	4.0	4.2	4.8	7.8	8.0	7.0	6.1	7.7	6.1	4.7
C_2H_6 do	.0	.0	.0	.0	.0	.0	.0	.0	.0	.0	.0	.0	.0	.0	.0	.0	.0
N_2 do	4.9	4.2	4.6	4.2	4.0	4.1	3.7	4.2	3.8	2.8	3.8	2.8	2.9	6.1	7.1	6.9	7.2

[1] Tyler standard screen-scale sieves.
[2] See table 8 for analysis of chars, table 11 for coal analysis, and table 12 for screen size.

PURIFICATION OF GAS

One of the important reasons for arriving at the decision to investigate gasification under pressure, such as was being done with the Lurgi generator, was the knowledge that a gas can be produced that has a higher heating value than gas made from coal by any other known method of complete gasification. It was hoped that a gas approaching 525 B. t. u. per cubic foot could be produced directly from coal, since the constantly increasing price of oil products made enrichment with oil economically unsound in competition with other fuels. In view of earlier results obtained with the generator, it became apparent that the Lurgi gas would have to be enriched or converted by some means to make a gas with a heating value of 525 B. t. u. per cubic foot. Since oil is expensive and the future supply limited, it was decided that part of the Lurgi gas should be synthesized to methane and mixed with the remaining gas to make a final product of 525 B. t. u. per cubic foot. A search of past and current literature revealed that the most promising methanization catalyst was the nickel catalyst being developed for the manufactured-gas companies in England by the Gas Research Board. This development has reached the pilot-plant stage. The first of a series of catalysts tested by the Board which showed promise in the laboratory was to be tested in the pilot plant.[11 12] However, by the time the pilot plant was ready for operation, a second catalyst had been developed that showed better resistance to carbon deposition and did not fail after testing for a period of 3,500 hours.[13 14 15]

The first problem that has to be solved in setting up a synthesizing unit using nickel as the main constituent in the catalyst is purification of the gas, particularly removal of the sulfur compounds. The first step taken to purify the Lurgi gas was construction of a pressure water-scrubbing tower which removed a large percentage of the CO_2 and H_2S from the gas. The tower was made from a 7-foot length of 6-inch pipe and filled with Berl saddles. Tests on this tower showed that, at a gas rate of 300 cubic feet per hour, about 90 percent of the CO_2 in the gas can be removed at 300 pounds per square inch pressure. The H_2S was

[11] Council of the Gas Research Board, Sixth Annual Report, 1944–45: Com. GRB 18, pp. 18–32.

[12] Dent, F. J., Moignard, L. A., Eastwood, A. H., Blackburn, W. H., and Hebden, D., An Investigation into the Catalytic Synthesis of Methane for Town Gas Manufacture; 49th Report of the Joint Research Committee of the Gas Research Board and the University of Leeds: Com. GRB 20, 1945.

[13] Council of the Gas Research Board, Seventh Annual Report, 1945–46: Com. GRB 24, pp. 20–34.

[14] Council of the Gas Research Board, Eighth Annual Report, 1946: Com. GRB 30, pp. 19–26.

[15] Gas Research Board, Report of the Director for the Year 1947: Com. GRB 37, pp. 10–19.

TABLE 10.—*Purification test data for Lurgi-process gas*

Test No.	57.	58.	59.
Date	12–4–47.	12–11–47.	12–17–47.
Time	12:40 p. m. to 3:00 p. m.	12:55 p. m. to 3:00 p. m.	12:40 p. m. to 2:25 p. m.
Fuel	No. 316 char.	No. 316 char.	No. 316 char.
Amount	33 lb.	28 lb.	26.5 lb.
Oxygen	196 cu. ft.	166 cu. ft.	148 cu. ft.
Steam	68.7 lb.	65.4 lb.	47.2 lb.
Gas	855 cu. ft.	700 cu. ft.	750 cu. ft.
Pressure	300 lb. per sq. in.	300 lb. per sq. in.	300 lb. per sq. in.
Temperature of purifier	300° C.	300° C.	200° C.
Space velocity	262.	212.	308.

GAS ANALYSIS, PERCENT

	After generator	After scrubber	After purifier	After generator	After scrubber	After purifier	After generator	After scrubber	After purifier
CO_2	26.2	11.5	20.3	25.8	13.6	17.2	27.6	7.0	13.2
Ill	.9	.7	.6	.3	.3	1.4	.6	.5	1.0
O_2	.1	.1	.4	.1	.3	.5	.4	.7	.5
H_2	42.7	49.4	45.5	42.8	53.3	45.7	41.8	53.9	48.5
CO	18.2	23.9	16.4	20.4	20.6	19.1	20.0	26.7	21.2
CH_4	8.3	10.2	12.8	8.1	9.5	11.7	5.9	7.3	11.4
C_2H_6	.0	.0	.0	.1	.2	.0	.0	.0	.0
N_2	3.6	4.2	4.0	2.4	2.2	4.4	3.7	3.9	4.2
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0

	Test 57	Test 58	Test 59
Sulfur in raw gas, grains per 100 cubic feet:			
H_2S	370	370	370
Organic	10	10	10
Organic sulfur after purifier, grain per 100 cu. ft.	0.51	0.29	0.31

TABLE 10.—*Purification test data for Lurgi-process gas*—Continued

Test No.	60.	61.	63.
Date	1-15-48.	1-27-48.	2-10-48.
Time	12:40 to 2:45 p. m.	12:40 to 2:45 p. m.	12:35 to 2:30 p. m.
Fuel	No. 268 char.	No. 316 char.	No. 316 char.
Amount	40 lb.	30.5 lb.	27.5 lb.
Oxygen	170 cu. ft.	170 cu. ft.	160 cu. ft.
Steam	68 lb.	67 lb.	65 lb.
Pressure	300 lb. per sq. in.	300 lb. per sq. in.	300 lb. per sq. in.
Gas	603 cu. ft.	637 cu. ft.	548 cu. ft.
Gas synthesized	137 cu. ft.	150 cu. ft.	172 cu. ft.
Temperature of purifier	200°-260° C.	180°-200° C.	180°-200° C.
Space velocity		160.	208.

GAS ANALYSIS, PERCENT

	After generator	After water wash	After purifier	After synthesis, CO_2-free	After generator	After water wash	After purifier	After synthesis, CO_2-free	After generator	After water wash	After purifier	After synthesis, CO_2-free
CO_2	27.8	2.9	12.3	0.0	24.8	4.2	1.9	0.0	28.6	19.6	19.7	0.0
Ill	.3	.3	.4	.4	.2	.3	.2	.8	.2	.4	.2	.6
O_2	.2	.5	.1	.2	.1	.2	.2	.2	.1	.1	.2	.2
H_2	41.9	55.8	50.6	.9	41.3	53.7	54.5	2.1	42.6	48.9	48.9	1.7
CO	18.3	26.3	15.2	.4	22.5	28.5	29.6	.7	18.4	20.4	20.3	.2
CH_4	6.3	8.9	17.7	92.8	7.6	9.5	9.8	94.5	7.1	7.6	7.7	91.8
C_2H_6												
N_2	5.2	5.3	3.7	5.3	3.5	3.6	3.8	1.7	3.0	3.0	3.0	5.5
Total	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0	100.0

Not determined.	Sulfur in raw gas, grains per 100 cubic feet: H_2S 370 Organic 10	Sulfur in raw gas, grains per 100 cubic feet: H_2S 370 Organic 10
Organic sulfur after purifier, 0.45 grain per 100 cu. ft.	Organic sulfur after purifier, 0.30 grain per 100 cu. ft.	Organic sulfur after purifier, 0.36 grain per 100 cu. ft.

usually reduced from about 350 grains to 6–10 grains per 100 cubic feet of gas. Removal of the organic sulfur presented a more difficult problem. Since the Germans have been using a soda-iron oxide catalyst for purifying their Fischer-Tropsch gases, it was decided to test this catalyst first. Two towers were made from 10-inch pipe, 36 inches long, and their screen trays filled with layers of catalyst of ½- by ¼-inch size, made from anhydrous soda ash and Masabi iron ore. A second batch of catalyst was made using Luxmasse, the iron oxide residue remaining in the purification of bauxite ores, instead of iron ore. Both catalysts seemed to work equally well. At 220° C. and above, the catalyst not only removed sulfur but synthesized some methane and carbon dioxide as well. At 180° to 200° C. the amount of sulfur removed depended on the space velocity—space velocity being the ratio of volume of gas (S. T. P.) passed per unit time to that of the bulk volume of the particles of the catalyst. The cleanest gas made had 0.3 grain of sulfur per 100 cubic feet when operating at a space velocity of 200. This concentration is probably too high to be tolerated by a nickel catalyst, and means of further purification of the gas are being considered. Table 10 shows the results of the gasification tests in which part of the gas was purified and synthesized. Coal 316, from which the char was made for some of the tests, is an Alabama coal from the Corona seam. The analysis of this coal is given in table 11 and the plastic properties in table 13.

TABLE 11.—*Analyses of coals, as-received basis* [1]

Coal No.	Seam and mine	Proximate, percent				Ultimate, percent					Ash fusion, °F.			Heating value, B. t. u. per lb.
		Moisture	Volatile matter	Fixed carbon	Ash	Hydrogen	Carbon	Nitrogen	Oxygen	Sulfur	Initial deformation	Softening	Fluid	
184	Anthracite	5.0	5.6	76.4	13.0					0.5				11,890
274	Semianthracite	2.7	8.8	65.7	22.8	3.1	67.2	1.4	4.8	.7	2,910+			11,280
294	Pittsburgh bed, Clyde mine	3.1	35.7	54.6	6.6	5.5	76.4	1.6	8.6	1.3		2,500		13,650
307	Disco	1.7	19.7	67.0	11.6						2,130	2,230	2,430	
316	Corona No. 20, Warrior Field, Ala	3.1	38.7	47.9	10.3	5.4	70.7	1.6	9.4	2.6	2,070	2,280	2,430	12,850
318	Anthracite, Barley	2.0	7.6	77.6	12.8	2.6	79.1	1.0	3.8	.7	2,890	2,910+		
319	Anthracite, Buckwheat No. 4	2.5	7.8	78.4	11.3	2.6	79.6	1.0	4.8	.7	2,840	2,910+		
320	Anthracite, Buckwheat No. 5	2.6	7.5	81.5	8.4	2.8	82.3	1.0	4.8	.7	2,910+			
367	Utah Coal, Hiawatha	4.8	45.7	46.2	3.3	6.4	74.2	1.4	14.1	.6	1,920	1,970	2,180	13,490
368	Jamison Coal, Pittsburgh bed	1.7	36.2	54.1	8.0	5.2	75.9	1.4	8.6	.9	2,310	2,340	2,490	13,560
397	Upper Freeport Coal, Kent No. 7	2.3	27.6	57.0	13.1	4.7	72.7	1.3	6.2	2.0	2,230	2,470	2,590	13,010
411	High-temp. coke, Brushton Coal Co.	6.7	1.4	80.7	11.2	1.1	79.4	.9	.3	1.3	2,150	2,380	2,540	11,590

[1] Analyses made under the supervision of H. M. Cooper, chemist, Central Experiment Station, Bureau of Mines, Pittsburgh, Pa.

TABLE 12.—*Screen analyses*

Screen Sizes

[Tyler Standard screen-scale sieves]

ANTHRACITE, COAL 184

	Percent
On 8-mesh	0.0
Through 8- on 10-mesh	.5
Through 10- on 14-mesh	6.9
Through 14- on 20-mesh	29.1
Through 20- on 35-mesh	51.1
Through 35- on 80-mesh	10.7
Through 80- on 100-mesh	.4
Through 100- on 200-mesh	.4
Through 200-mesh	.9
	100.0

ANTHRACITE—BARLEY, COAL 318

	Percent
On 4-mesh	3.8
Through 4- on 6-mesh	27.2
Through 6- on 8-mesh	31.6
Through 8- on 10-mesh	23.3
Through 10- on 14-mesh	8.7
Through 14- on 20-mesh	3.4
Through 20- on 35-mesh	1.5
Through 35- on 60-mesh	.3
Through 60- on 100-mesh	.1
Through 100-mesh	.1
	100.0

SEMIANTHRACITE, COAL 274

	Percent
On 6-mesh	66.7
Through 6- on 8-mesh	13.3
Through 8- on 10-mesh	9.6
Through 10- on 14-mesh	6.1
Through 14- on 20-mesh	2.9
Through 20- on 35-mesh	.7
Through 35- on 80-mesh	.2
Through 80- on 100-mesh	.1
Through 100- on 200-mesh	.1
Through 200-mesh	.3
	100.0

ANTHRACITE—BUCKWHEAT NO. 4, COAL 319

	Percent
On 8-mesh	0.3
Through 8- on 10-mesh	6.2
Through 10- on 14-mesh	23.7
Through 14- on 20-mesh	33.6
Through 20- on 35-mesh	28.9
Through 35- on 60-mesh	4.8
Through 60- on 100-mesh	1.1
Through 100-mesh	1.4
	100.0

ANTHRACITE—BUCKWHEAT NO. 5, COAL 320

	Percent
On 8-mesh	0.2
Through 8- on 10-mesh	1.3
Through 10- on 14-mesh	8.3
Through 14- on 20-mesh	35.8
Through 20- on 35-mesh	47.6
Through 35- on 60-mesh	4.0
Through 60- on 100-mesh	.9
Through 100-mesh	1.9
	100.0

TABLE [illegible].—*Plastic properties of coal*

Coal No.	Fusion temperature, Davis plastometer, resistance develops, °C.	Maximum fluidity, Davis plastometer, minimum resistance		Solidification, Davis plastometer, maximum resistance		Davis plastometer resistance ends, °C.
		°C.	Lb.-in.	°C.	Lb.-in.	
316	396	428–437	0.9	469	11.9	482
367	No resistance developed at normal rate of heating of 3° C. per minute.					
368	398	418–42	.0	463	11.3	499
397	390	426–45	.0	495	40.5	528

SYNTHESIS TESTS

The nickel catalyst first tried in a laboratory-scale converter was developed by the Gas Research Board in England and reported in its publications GRB 20 and 24.[16 17] The catalyst chamber consisted of a ½-inch seamless pipe, 25 inches long, jacketed with a 2½-inch pipe, 15 inches long. The catalyst tube was heated externally with boiling Dowtherm in the jacket. In this manner, it was possible not only to maintain a constant temperature but to obtain any operating temperature up to 400° C. in the catalyst by simply pressurizing the jacket. The Dowthern jacket was heated electrically by a resistance winding on the outside of the 2½-inch pipe and insulated with magnesia pipe covering. The vapors were condensed in a water-cooled condenser connected to the jacket. When mixtures of bottled H_2 and CO gases were synthesized at 300 pounds per square inch pressure, the heat of reaction was partly dissipated by the Dowtherm and partly carried out by the effluent gas. Usually, the space velocity of the gases was increased until the effluent gas had a temperature of about 450° C.

The catalyst, made by precipitating 100 parts by weight of nickel nitrate, 20 parts of manganese nitrate, 13.6 parts of aluminum nitrate, and 25 parts of china clay with 97 parts of potassium carbonate, was crushed to 8- by 12-mesh (Tyler standard sieves) and placed in the catalyst chamber. Four thermocouples, 3 inches apart, were inserted in the catalyst and connected to a recording potentiometer. The catalyst was reduced for 2 hours with hydrogen at 400° C. and 300 pounds per square inch pressure.

The first synthesis tests were more or less exploratory in character, and no reliable data were obtained. Table 14 gives the results of two tests made to determine the activity of this catalyst. In synthesis tests 7 and 8, the required conditions were obtained by varying the ratios of H_2 : CO and taking samples of the effluent gases. Each test took about 6 hours to complete. In synthesis test 7 the H_2 : CO ratio was varied, while the space velocity was held approximately constant. In synthesis test 8 the H_2 : CO ratio was held virtually constant, while the space velocity was varied. In all instances, the reaction was rapid enough to reach approximate equilibrium in the 12-inch layer of catalyst. The temperature of the effluent gas ordinarily rose above the 450° C. recommended by the Gas Research Board, and the CO_2 in the gas would indicate that carbon was being deposited by the reaction $2\ CO = C + CO_2$. No attempt was made to measure the life or the effects of poisons on the catalyst. After synthesis test 8, the catalyst chamber was opened and the catalyst examined. Instead of being in 8- by 12-mesh size, as originally charged, it now contained some powder, black in appearance, indicating that

[16] See footnote 12.
[17] See footnote 13.

TABLE 14.—*Laboratory tests on synthesis catalyst*

Ratio H_2 : CO	Space velocity[1]	Dowtherm, °C.	Analysis of effluent gas, percent				Temperature of effluent gas, °C.	Remarks
			CO_2	H_2	CO	CH_4		
4.3	5, 000	300	0. 0	58. 8	0. 2	34. 6	390	Synthesis test 7.
4.0	5, 000	300	. 0	53. 6	. 4	43. 7	432	Do.
3.75	5, 500	300	. 0	46. 7	. 4	50. 1	452	Do.
3.5	6, 000	300	. 0	37. 6	. 6	55. 7	480	Do.
3.1	5, 500	300	. 0	25. 7	. 4	68. 5	482	Do.
3.0	6, 000	300	. 2	11. 7	. 5	80. 8	500	Do.
2.7	[illegible], 500	300	1. 4	8. 0	. 6	80. 5	494	Do.
3.2	5, 500	300	. 0	23. 2	. 4	70. 9	516	Synthesis test 8.
3.2	6, 200	300	. 0	27. 2	. 6	68. 0	471	Do.
2.9	6, 600	300	3. 5	16. 5	. 4	73. 5	483	Do.
3.0	10, 100	300	3. 9	17. 3	. 2	69. 6	513	Do.
3.4	15, 100	300	1. 0	25. 8	. 0	66. 9	510	Do.
3.0	21, 200	300	5. 0	15. 6	. 4	73. 1	574	Do.

[1] Space velocity $= \frac{V}{v}$, where V=cu. ft. of feed gas per hour and v=cu. ft. of catalyst (bulk volume).

an appreciable amount of carbon was deposited during the tests. The laboratory tests, which were so far carried out with bottled gases, were discontinued after it was ascertained that the catalyst was as active as described by the Gas Research Board.

To synthesize the gas from the Lurgi generator, a larger converter was built. Ninety-two tubes of ½ inch inside diameter were welded at the ends into two headers and then jacketed with a 12-inch pipe. The Dowtherm was again heated by a resistance winding around the jacket. Two water-cooled condensers were connected to the jacket to condense the Dowtherm vapors. This larger converter was charged with catalyst, assembled, and placed in the gas train following the iron oxide-soda purifier vessels (see fig. 7). In table 10, tests 60, 61, and 63 show the analyses of the synthesized Lurgi gases on a CO_2-free basis. It will be observed that, on this basis, the heating value of the synthesized gas (mostly methane) is over 900 B. t. u. In view of these results, the possibility of making high B. t. u. pipeline gas as a substitute for natural gas was considered.

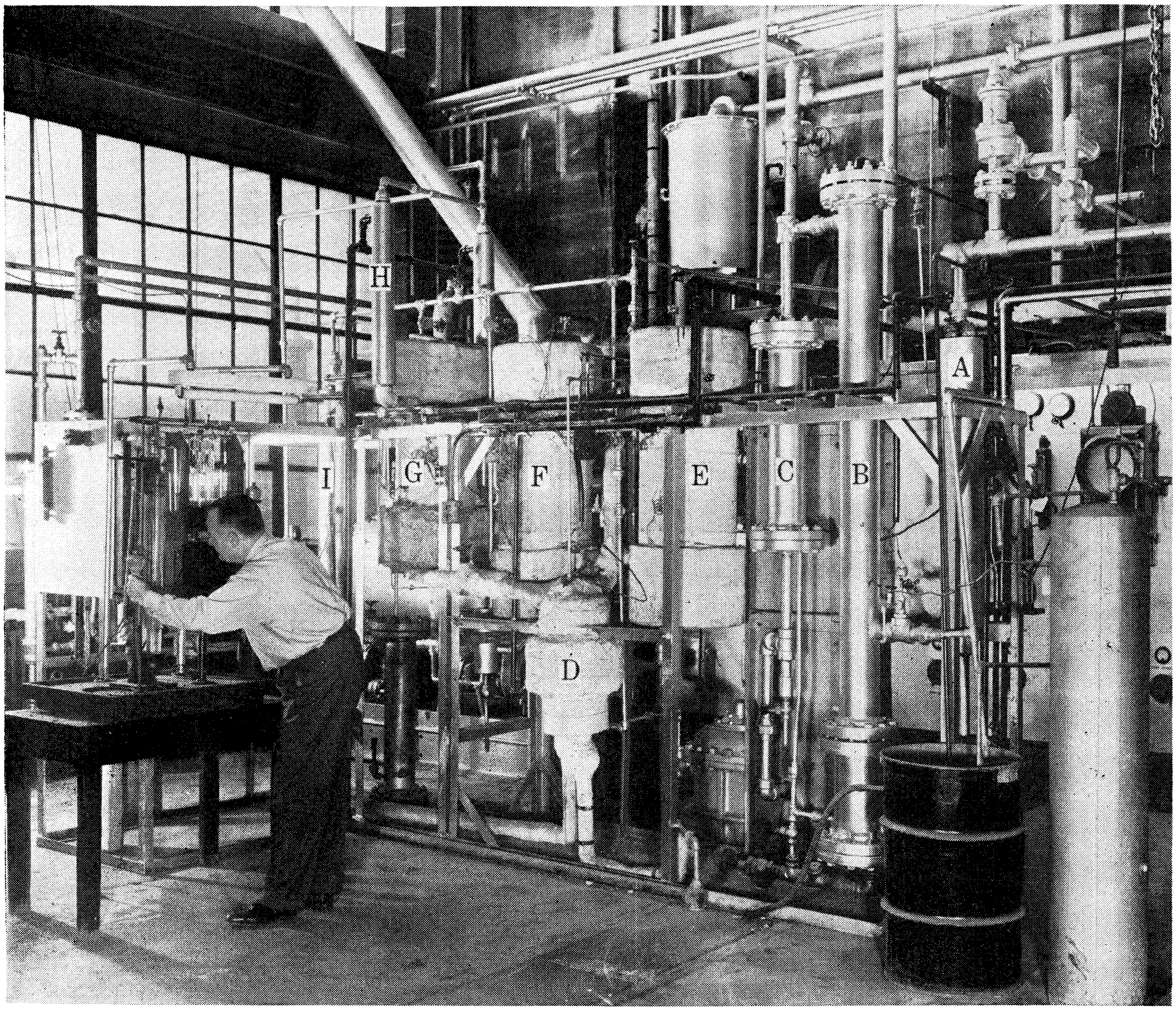

FIGURE 7.—PURIFICATION AND SYNTHESIS TRAIN.

A, Indirect Condenser; *B*, Cooler Scrubber; *C*, Water Trap; *D*, Gas Heater; *E*, Purifier I; *F*, Purifier II; *G*, Converter; *H*, Dowtherm Condenser; *I*, Indirect Cooler.

DESCRIPTION OF 13.5-INCH CONTINUOUS GENERATOR

The results of about 60 tests made to date with the Lurgi generators have indicated that it is technically feasible to gasify both treated bituminous coals and anthracites under pressure. However, neither the design of a commercial generator from an engineering viewpoint, nor the cost of producing pipeline gas from an economic viewpoint, could be ascertained from the batch pilot-plant tests. In the reports received from Germany, stress is laid on the ash-fusion point of the coals to be gasified. Apparently, coals with low ash-fusion temperatures caused clinkering trouble in the generator and could only be successfully gasified at low rates and with low efficiencies. In the batch generators used in the present work, no ash was removed during the tests; consequently, no clinkering problem was encountered. Also, the same German sources report capacities that seem to be extremely small for pressure gasification, as compared with nonpressure gasification methods. Finally, in any economic calculations, the purification and conversion costs would be an important item. The life of the catalyst could not very well be determined with a generator that can only operate 3 hours per test. In view of these considerations, it was decided to build a third generator that could be operated continuously.

Dr. O. Hubmann, formerly of the German Lurgi company, was available at the Central Experiment Station during the spring of 1947. He designed a continuous Lurgi generator of 1 square foot gasification area for the Bureau of Mines. The engineering and construction of the generator were handled by the Blaw-Knox Corp. in Pittsburgh, Pa. The generator proper consists of a heavy steel cylinder, 5 feet 5 inches high and 20 inches outside diameter, which is welded top and bottom to suitable flanges. To the inner wall of the cylinder is welded at top and bottom a second steel cylinder, 16 inches outside diameter, with collars on the ends to form a water jacket. A stainless steel liner, exactly 13.5 inches inside diameter and ¼ inch thick, is set off from the water jacket by a ½-inch air space. The liner is held in place by a flange, welded on its lower end, between the cylinder and grate housing flanges. A ½-inch opening above the water level in the water jacket connects to the gas outlet so that the pressure in the generator is carried only by the outside cylinder. The gasification of the coal takes place within the liner, and, since it is ½ inch from the inner wall of the water jacket, excessive heat losses through a water-cooled wall are eliminated. The gas offtake is in the upper part of the shell. Directly beneath the liner is the rotating grate on which the charge rests and through which the oxygen-steam mixture enters. The ash and charging hoppers are each provided with two valves for discharging ashes and charging fuel while operating under pressure. The fuel bin, which holds approximately ½ ton of material, feeds into the charging hopper. The oxygen-steam mixture is superheated electrically. The oxygen is obtained from a "cascade" bank provided by the Linde Air Co., and has a capacity of 20,000 cubic feet of oxygen. The steam is obtained from a 15-horsepower, 500-pound-per-square-inch boiler from the Cyclotherm Corp. A recording potentiometer records temperatures (1) of the incoming oxygen-steam mixture before the superheater, (2) after the superheater, (3) at the grate, (4, 5, and 6) of the outer surface of the stainless-steel liner at various distances up from the gate, (7) of the effluent gas, and (8) of the gas after the cooler. In addition, the panel board includes meters for measuring the rates of flow of steam, of oxygen, and of the gas produced, a recording meter for measuring the CO_2 in the effluent gas, a recording gas calorimeter, and a back-pressure regulating pilot. Figure 8 shows the generator without the fuel bin, which extends through the roof of the building, a part of the panel board, the condenser, and the washing tower of the synthesis train. The batch generator shown at the right is also connected to the synthesis train and was used in making preliminary tests on new coals. Figure 9 pictures the complete panel board and high-pressure steam boiler. Figure 10 is the flow diagram of the Lurgi pilot plant.

A test is started by heating a pail of char to red heat and then pouring it in the generator through the charging hopper, the two valves of which are left wide open. A second pail of cold char is added and the generator allowed to heat. A low-pressure air pump is throttled down so that about 400 cubic feet of air per hour enters the generator through the superheater held at 150° C. When the liner temperature indicates that gasification is proceeding satisfactorily, more charge is added. The generator is fully charged and the valve to the coal hopper closed in about 1 hour after the initial charge of hot char. Meanwhile, the temperature in the generator is controlled by saturating the influent air with steam, so that

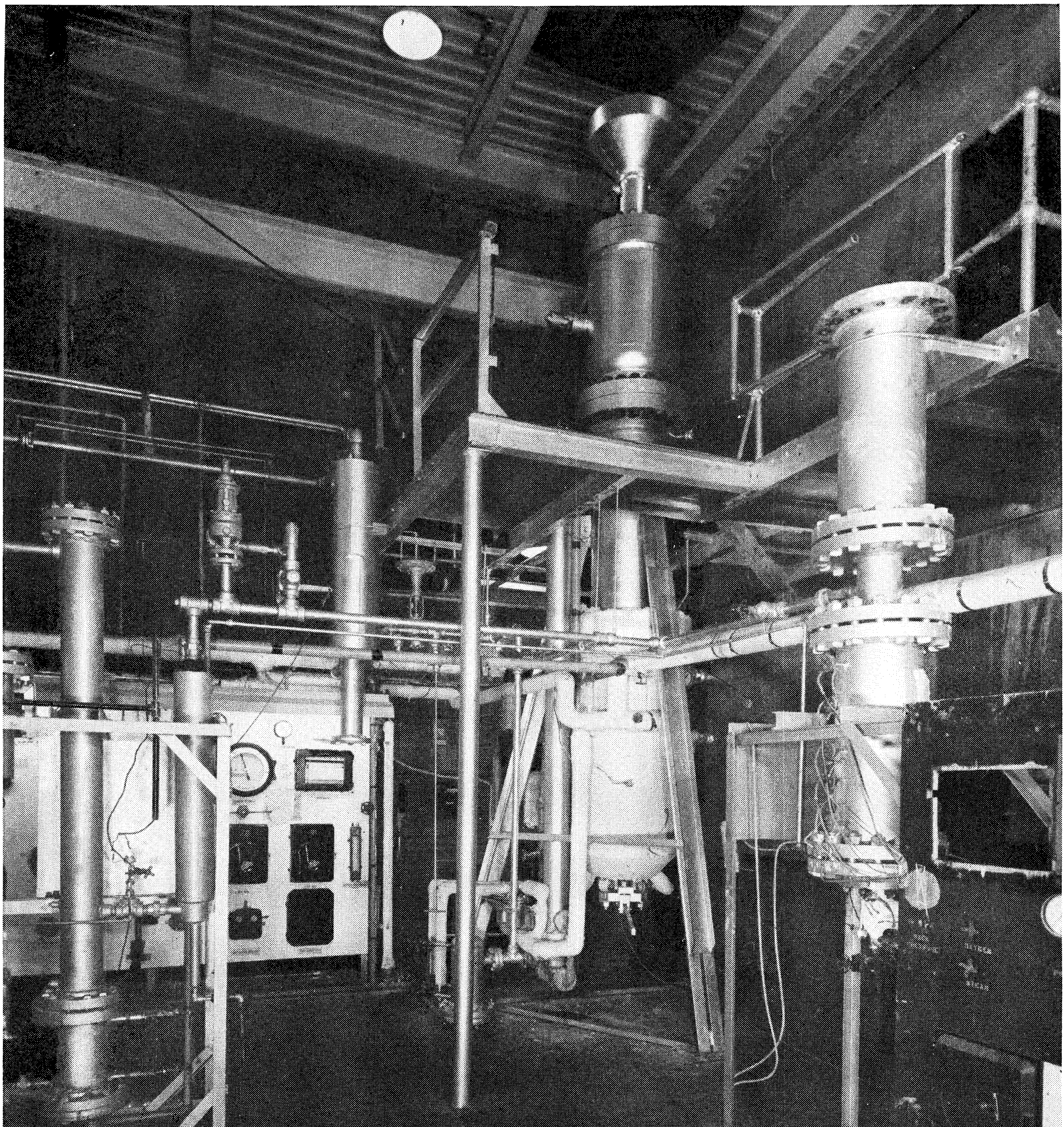

FIGURE 8.—13.5-INCH, HIGH-PRESSURE, LURGI CONTINUOUS GENERATOR.

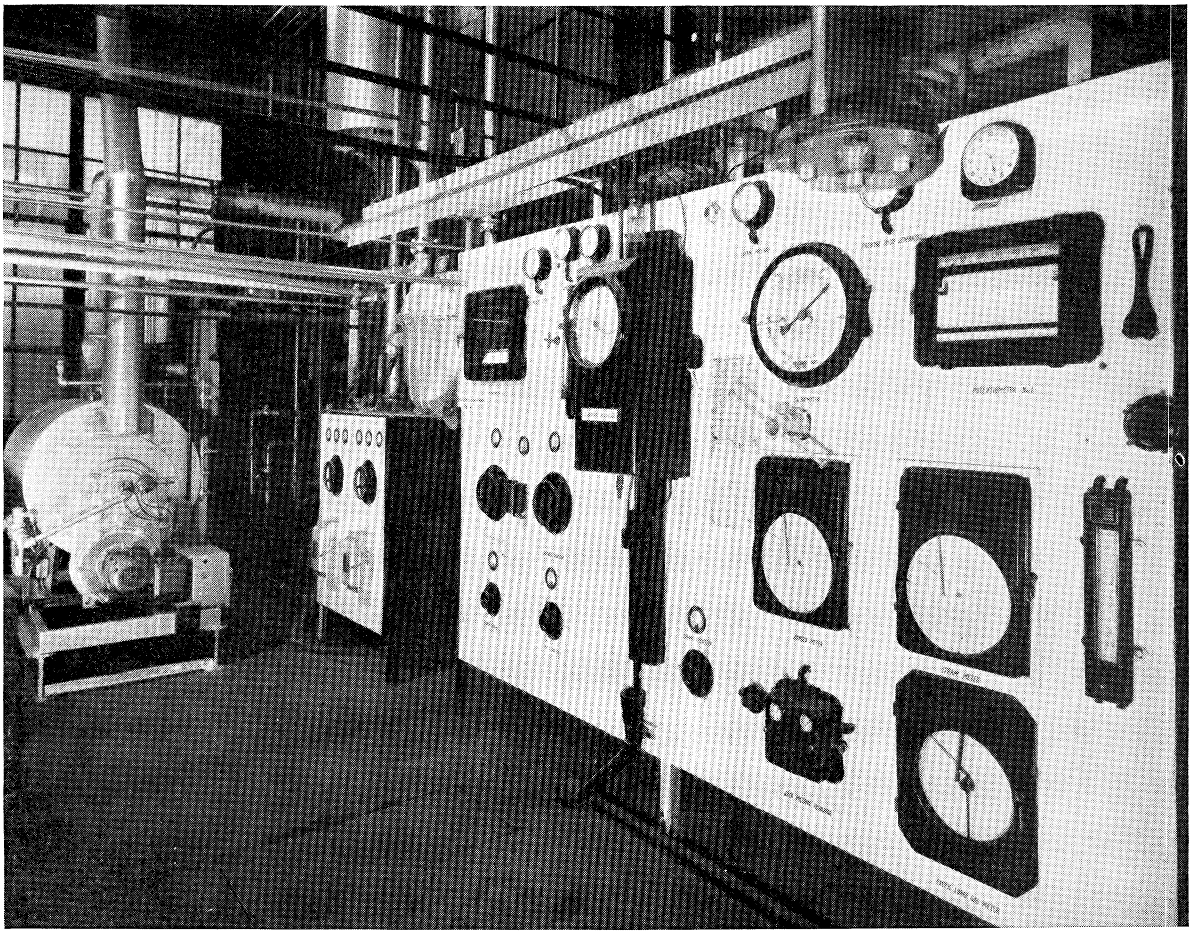

FIGURE 9.—PANEL BOARD AND STEAM BOILER.

the temperature of the liner remains at about 500° C. It was found by experience that, when the air is heated and saturated with steam to 70° C., the liner temperature remains below 500° C. When the temperatures indicate that a steady state has been reached, the saturated air is replaced with the steam-oxygen mixture. The temperature of the liner determines the ratio of oxygen to steam. The pressure-regulating pilot is set to increase the pressure in the generator by 50-pound increments. After each increment the superheater temperature is increased and the generator permitted to reach a steady state of operation. Meanwhile, the CO_2 recorder has been placed in operation. When the operating pressure has been reached, the settings for the desired test conditions are made, and the test is continued to obtain the required data. When in operation, it has been found that the progress of the test can best be regulated from the amount of CO_2 in the effluent gas, the liner temperature being used only to determine the position of the gasification zone. The speed of the ash extractor, which is connected to a ½-horsepower motor through a reducing gear, is determined by the change in position of the hot zone in the liner. When the gas leaves the generator, it passes through an indirect precooler, also provided with a water spray for direct cooling. The gas pressure is reduced through a back-pressure regulating valve, and the gas is washed with a water spray. It is then sampled, metered, vented, and burned in the air. When a synthesis test is made, a part of the Lurgi gas is piped *under pressure* through the synthesis train; then the pressure is reduced through a back-pressure-regulating valve, after which the gas is sampled, metered, vented, and also burned in the air. The water and tar, separated from the gas in the precooler, are removed through an automatic valve controlled by a water-level float. A more detailed description and the operation of a test (No. 86) are given in the appendix.

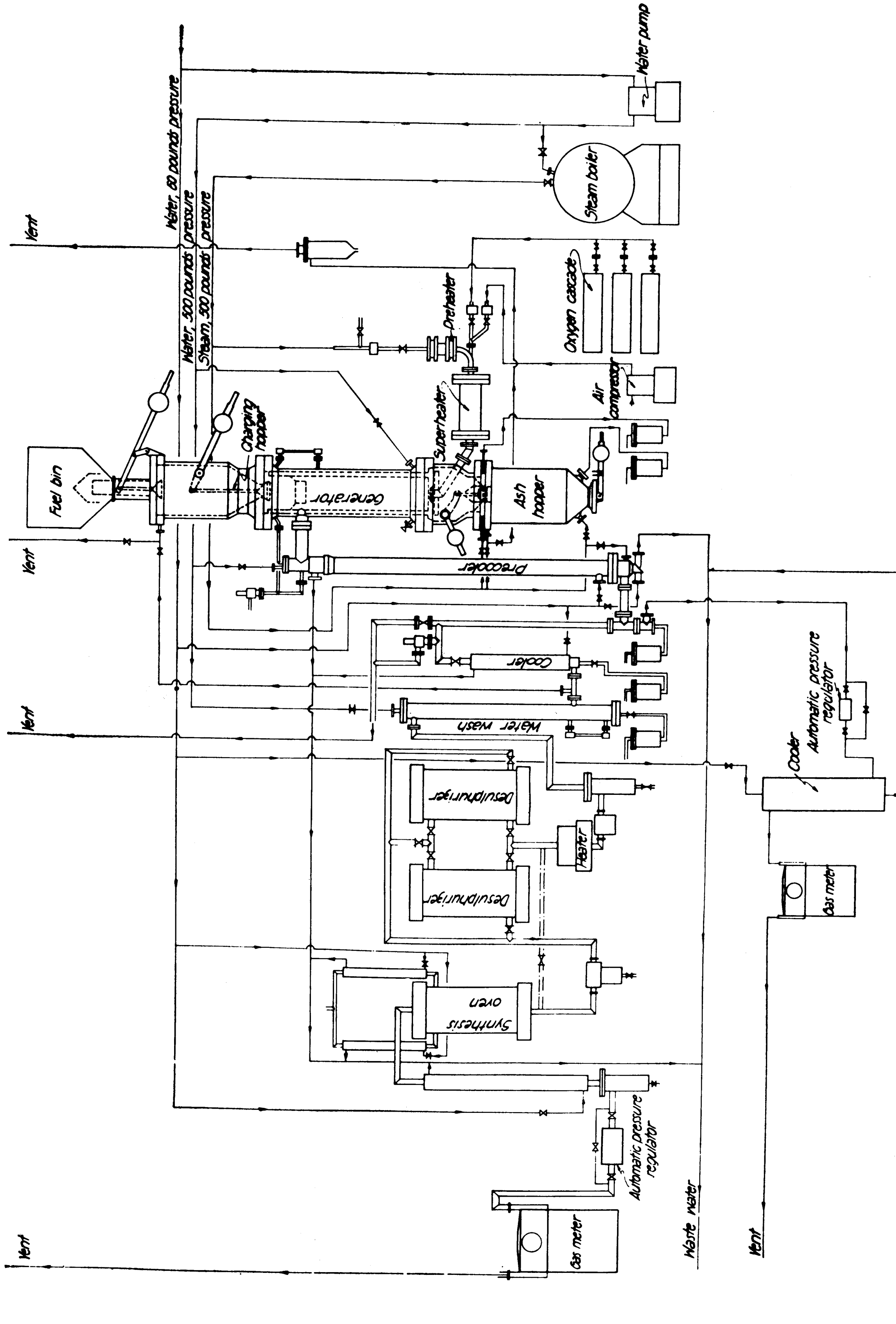

FIGURE 10.—FLOW DIAGRAM OF LURGI PILOT PLANT.

DESCRIPTION OF ROTATING RETORT

To make char for the continuously operating generator, a rotary batch retort of ½-ton capacity was designed and built. The retort is a closed cylinder, 8 feet long and 4 feet in diameter, made from ¼-inch steel plate. Through the center and extending about 2 feet out of both ends is a 6-inch pipe used as a horizontal shaft, on which the retort is rotated. The shaft is mounted at both ends in roller bearings on supports 5½ feet tall. The retort is heated on the bottom with natural gas. A shell made from ⅛-inch steel plate covered with asbestos blocks is placed over the retort to insulate it. The retort is driven by a sprocket and chain to an electric motor and provided with a gear arrangement to vary the speed. The gas, tar, and liquor produced during the carbonization of the coal escape through holes drilled in the 6-inch pipe and pass through a stuffing box into the purification train. The products are cooled in an indirect, water-cooled, tubular condenser, then in a second single-tube condenser. The gas is further cleaned by a Cottrell precipitator and finally metered. The products were measured and sampled for analyses. The results for the four bituminous coals carbonized are given in table 15.

TABLE 15.—*Results of carbonization tests*

Coal	Corona 316.	Hiawatha 367.	Pittsburgh 368.	Upper Freeport 397.
Carbonization time (starting with cold retort)	5 hr.	4 hr.	6½ hr.	5 hr.
Charge	1,000 lb. coal + 180 lb. fines.	525 lb. coal.	1,000 lb. coal + 300 lb. fines.	900 lb. coal + 300 lb. fines.
Products recovered, pounds				
Total char	958.0	397.0	1,123.0	1,055.0
Tar (wet)	114.0	74.6	120.0	65.5
Water	68.5	41.6	18.0	42.5
Gas (850-900 B. t. u.)	39.5	11.8	39.0	37.0

TABLE 15.—*Results of carbonization tests*—Continued

Coal	Corona 316.	Hiawatha 367.	Pittsburgh 368.	Upper Freeport 397.
Char analysis, percent				
Volatile matter	23.1	22.9	20.9	14.1
Fixed carbon	64.1	68.5	68.4	70.0
Ash	12.8	8.6	10.7	15.9
Wet tar analysis, percent				
Dry tar	82.93	83.3	80.2	79.1
Water	17.07	16.7	19.8	20.9
Dry tar analysis, percent				
Pitch	41.4	49.6	44.3	52.6
Neutral oil	34.7	37.0	34.0	33.8
Acids	22.1	11.0	20.4	12.4
Bases	1.8	2.4	1.3	1.2
Solids	.0	.0	.0	.0
Dry tar, specific gravity				
Specific gravity, $\frac{15.6\ °C.}{15.6\ °C.}$	1.003	0.972	1.012	1.047
Dry-tar distillation, percent				
Fraction and range:				
1. −170° C.	6.9	4.9	8.1	5.5
2. 170°-235° C.	18.7	13.9	17.8	16.4
3. 235°-270° C.	10.5	9.9	11.2	8.3
4. 270°-350° C.	22.5	21.7	18.6	17.2
Pitch	41.4	49.6	44.3	52.6
Recovery per ton of coal charged				
Char	1,556 lb.	1,515 lb.	1,646 lb.	1,678 lb.
Tar (dry)	22.8 gal.	28.5 gal.	23.1 gal.	13.5 gal.
Gas	1,109 cu. ft.	630 cu. ft.	1,090 cu. ft.	1,143 cu. ft.

RESULTS WITH CONTINUOUS GENERATOR

Tests 64 to 79 were made to determine operating characteristics, and complete data were not taken. The generator was ready for the first test (64) on June 15, 1948. Minor troubles in the piping and generator caused the test to be discontinued after 8 hours. In the next two tests (65 and 66), made with Corona char, the grate did not remove ashes, and the tests had to be stopped when the combustion zone moved up in the generator toward the gas outlet. During test 67, made with Barley anthracite, the stainless-steel liner was burned through in several places and had to be replaced. In the next five tests, various changes in the grate and in operation of the generator were tried, with the result that test 73 was successfully completed. The gas rate in this test was about 2,500 cubic feet per hour when operating under pressure. In tests 74 and 75 some difficulties were again experienced in removing ash. Stationary plows bolted to the generator just above the grate and extending 3 inches over it overcame this difficulty. Tests 76 and 77 were made in the batch generator with Hiawatha coal from Utah. The analysis and the plastic properties of this coal are given in tables 11 and 13. Although this coal has very poor coking properties, it coked solid in the generator under 300 pounds per square inch pressure.

Table 16 gives the results of the tests made after test 79. The operating time for the majority of the tests ranged between 20 and 27 hours. The data in table 16 are taken from that portion of the test after desired operating conditions were reached. Many of the tests up to No. 90 had some clinkers on the stationary plows. To overcome this, a stainless-steel, ½- by 1-inch bar, parallel to and 2 inches above the grate, was welded to an upright on the grate. The bar rotates with the grate and sweeps the top of the plows twice every revolution. In the remaining tests (90–97) no trouble was experienced with clinkers; the clinkers formed were crushed and removed by the grate.

Beginning with test 80 the main object was to test the char and to determine the capacity of the generator. It was found that the char of all coals acts similarly in the generator and that minor discrepancies could be accounted for by the difference in the ash. Therefore, after a char from a particular coal was tested to obtain such information as gas yield per ton of char, steam decomposition, and ash performance, the next char was tested, even though the maximum capacity of the generator had not been reached. In designing this generator, a maximum capacity of 6,600 cubic feet of raw gas per hour per square foot gasification area was assumed. In view of the results obtained and published in Germany, this capacity was considered to be the maximum obtainable under the most favorable working conditions. Accordingly, all of the accessory equipment was specified for producing, cleaning, cooling, and measuring 6,600 cubic feet of gas per hour. When test 90 indicated that rates above 7,000 cubic feet per hour could be obtained, the steam and oxygen flow orifices were enlarged, and arrangements were made to have each of two meters measure part of the gas. In test 92 the gas rate was slowly increased to 7,250 cubic feet per hour and held at this point until an hour or so before the end of the test, when the rate was increased to 9,000 cubic feet per hour. No trouble was experienced at this rate except that the boiler capacity was exceeded and the boiler pressure dropped from the normal operating pressure of 400 pounds per square inch to 360 pounds per square inch, when the test was stopped. Tests 93 (not shown in table 16) and 94 were made to obtain performance data on a coal from the Upper Freeport seam. Parts 94a and 94b of test 94 were made at pressures of 375 and 420 pounds per square inch, respectively. There was nothing unusual about these tests, even though the ash content of this char was higher than in chars previously tested. When the ash hopper was filled the test was discontinued, because the upper ash-discharge valve leaked under pressure. The present plans are to complete this program before repairing this valve.

It will be noted in table 16 that, as the capacity of the generator was increased, the methane in the gas decreased. Since the thermal efficiency in making pipeline gas depends on the methane synthesized inside the generator, some thought was given to the problem of maintaining or increasing the amount of methane synthesized in the generator at high capacity rates. Figure 11 shows the relation between the maximum efficiencies that can be expected in making a pipeline gas of about 950 B. t. u. with various amounts of methane in a theoretical Lurgi gas. Since all factors, except capacities, are similar in tests made with the same char, the methane synthesis in the generator must depend on the contact or reaction time between gas and char. At a high operating rate this contact time decreases; as a result, the methane in the gas decreases.

TABLE 16.—*Test results of operation of Lurgi pilot generator*

Test No.[1]	Fuel		Steam, lb. per			Oxygen, cu. ft. per 1,000 cu. ft. gas	Steam decomposition, percent	Gas production		Gas analysis, percent									Ratio H_2:CO	Calorific value		Superheater temperature, °C.	Gasification pressure, lb. per sq. in.
	Kind	Lb. per hr.	Hour	100 lb. fuel	1,000 cu. ft. gas			Cu. ft. per sq. ft. of generator area	Cu. ft. per ton of fuel	CO_2	Ill.	H_2S	O_2	H_2	CO	CH_4	C_2H_6	N_2		Raw gas	Washed to 2.5 percent CO_2		
Corona Seam Coal (Alabama), No. 316																							
80	Char---	91	231	255	66	158	35.2	3,483	77,000	29.45	0.5	0.65	0.2	39.7	16.3	11.4	0.1	1.7	2.4	312	438	332	300
81	---do---	103	229	223	58	142	39.1	3,942	76,800	28.2	.4	.65	.2	39.9	18.6	8.3	.85	2.9	2.1	295	414	328	300
82A	---do---	97	209	215	57	152	40.2	3,634	75,000	29.95	.4	.65	.0	40.0	16.6	10.2	.9	1.3	2.4	313	445	220	300
82B	---do---	101	217	215	55	151	41.2	3,936	77,700	28.35	.3	.65	.0	40.6	18.5	9.9	.4	1.3	2.2	307	427	212	300
Pittsburgh Seam Coal (West Virginia), Jamison Mine, No. 368																							
85A	Char---	98	203	212	54	158	38.3	3,740	78,300	26.8	0.5	0.4	0.0	41.1	20.5	8.7	1.3	0.7	2.0	321	432	212	300
85B	---do---	110	240	229	54	148	39.6	4,411	80,100	26.8	.4	.4	.0	41.2	21.4	7.9	1.1	.8	1.9	311	419	212	300
86	---do---	114	241	211	55	144	41.9	4,401	79,200	26.6	.5	.4	.0	41.4	19.8	9.0	1.3	1.0	2.09	320	425	315	300
90	---do---	146	352	241	59	136	40.2	6,017	83,600	28.6	.1	.2	.3	40.2	20.1	7.1	1.0	2.4	2.0	289	393	360	300
91	---do---									28.3	.4	.2	.4	39.5	20.3	8.6	.4	1.9	1.95	295	402	404	300
91a	---do---	} 98	261	267	65	136	35.8	4,029	82,500	29.0	.4	.2	.4	39.8	18.2	9.3	.6	2.1	2.19	300	412	404	375
91b	---do---									27.7	.2	.2	.4	40.1	18.4	8.4	1.4	3.2	2.18	304	409	404	405
92	---do---	177	499	282	79	127	33.6	7,246	82,100	29.2	.2	.2	.2	41.4	19.8	6.4	.7	1.9	2.09	281	386	420	300
Hiawatha Seam Coal (Utah), No. 367																							
88	Char---	94	242	257	65	146	38.2	3,746	79,600	31.4	0.6	0.04	0.2	40.4	14.9	9.66	0.9	1.9	2.71	302	429	350	300
89A	---do---	91	231	254	62	136	39.2	3,721	81,900	30.4	.5	.04	.2	42.1	15.8	8.56	.9	1.5	2.71	298	428	350	300
89B	---do---	118	303	258	64	144	35.8	4,717	80,100	28.5	.5	.04	.2	40.3	20.2	7.66	1.3	1.3	2.0	305	417	330	300
Upper Freeport Bed, Kent No. 7 Mine, No. 397																							
94	Char---	130	298	230	56	145	37.4	5,246	81,000	28.6	0.4	0.4	0.1	40.5	22.4	5.8	0.3	1.5	1.81	274	378	373	300
94a	---do---	130	298	230	56	145	37.4	5,246	81,000	26.6	.4	.4	.1	41.4	22.2	7.3	.4	1.2	1.87	293	391	373	375
84b	---do---	130	298	230	56	145	37.4	5,246	81,000	26.6	.4	.4	.1	40.4	22.6	7.9	.2	1.4	1.79	294	393	373	420
94[2]	---do---	----	----	------	------	------	------	------	------	32.4	1.2	.4	.1	32.7	26.2	4.1	1.1	1.8	1.25	271	393	373	300
94[2]	---do---	----	----	------	------	------	------	------	------	28.3	1.2	.4	.1	33.2	30.1	5.5	.1	1.1	1.10	281	385	373	300
94[2]	---do---	----	----	------	------	------	------	------	------	27.2	1.4	.4	.3	28.5	35.3	5.5	.0	1.4	.81	284	383	373	300
Pittsburgh Seam High-Temperature Coke, No. 411																							
96	Coke---	125	359	287	71	214	28.1	5,028	80,500	30.5	0.3	0.3	0.2	40.1	25.2	2.2	0.1	1.1	1.54	244	336	340	300
97	---do---	122	291	239	57	239	33.5	5,094	83,700	26.9	.3	.1	.1	38.6	31.1	2.3	.0	.6	1.24	253	335	310	300

[1] Test numbers followed by capital letters represent individual tests; test numbers followed by small letters represent a part of the numbered test during which special gas samples were collected for analyses and calorific value determinations.
[2] Carbon dioxide substituted for steam; volumes substituted not measured.

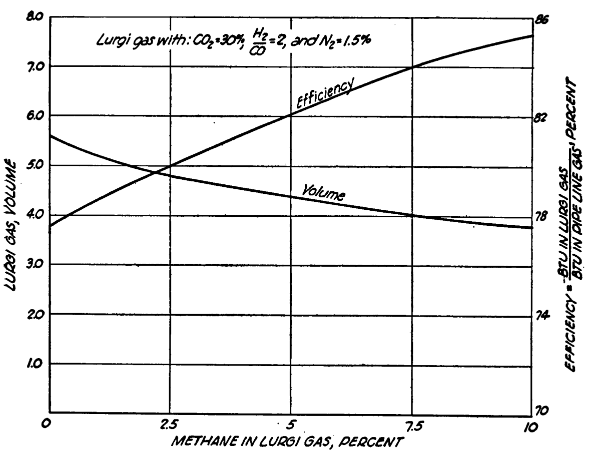

FIGURE 11.—LURGI GAS REQUIRED TO MAKE 1 CUBIC FOOT OF PIPELINE GAS.

There are two ways by which the contact time can be increased for a constant throughput: (1) Increase the height of the generator and (2) increase the operating pressure. The curve of methane content, however, flattens, and increase in methane will not be directly proportional to the increase in pressure. It appears, therefore, that the best way to increase the methane content of the Lurgi gas, without interfering with performance in other respects, is to increase the height of the generator. This modification would mean building a new generator unit and would require more headroom, which in turn would mean addition to the present building. The modification has not been attempted as yet.

Many variables, some of which have not been adequately investigated, are involved in passing through the Lurgi process and finishing with the pipeline high-B. t. u. gas, that it is difficult to give an analysis of the complete method consistent with maximum economy. However, perhaps an approximate estimate is appropriate: Basing the estimate on test 90, table 16, in which a moderate gasification rate was maintained, and assuming that 3,500 cubic feet of Lurgi gas are required to produce 1,000 cubic feet of pipeline gas (which has been obtained), there are also required:

59 x 3.5 =206 lb. of steam
136 x 3.5 =476 cu. ft. of oxygen
3,500 x 2,000 / 83,600 =83. 7 lb. of char

The analysis of the raw Lurgi gas is given in table 16. The pipeline gas, synthesized from a similar gas, analyzed (CO_2-free) as follows:

CO_2	Ill.	O_2	H_2	CO	CH_4	N_2	B.t.u. CO_2-free calculated	B.t.u. CO_2-free determined
0. 0	0. 6	0. 8	1. 9	0. 8	90. 7	5. 2	930	990

The design of this generator was based, in part, on the results of German practice. When gasifying brown coal or lignite, as was done in Germany, the required height of the generator was limited because of the high moisture content of these fuels. The relatively high temperature required for the synthesis of methane was quickly lowered in drying the charge. However, the rate of gasification in Germany was low—about 2,000 cubic feet per hour per square foot area; as a result, the methane content of the gas was comparatively high. In the continuous generator, used in the present work, the offtake-gas temperature increased from 400° C. to over 600° C. when the gasification rate was increased from 3,000 to 7,000 cubic feet per hour. An offtake temperature of 600° C. and above indicates that, if a higher generator were used, a higher methane content in the gas could be expected, since there are some indications that the methane reaction proceeds at a finite velocity at this temperature. In the present generator this high-temperature heat is dissipated in the coolers. Figure 12 illustrates the effect of temperature on the methane content of the Lurgi gas and indicates clearly that lower temperatures than those prevailing in the shaft of the present Lurgi set-up are desirable.

That a higher operating pressure increases the methane is shown in tests 94, 94a, and 94b, table 16. This may be due to the following reasons: (1) The char-gas reaction time is increased due to the slower travel of the gas through the generator, and (2) the equilibrium for the formation of methane is favored by increase in pressure. (See fig. 2.) By increasing the pressure from 300 to 420 pounds per square inch, the flow rate of the gas in the generator is decreased by about one-third. Since the rate of operation in test 94b was about 5,250 cubic feet per hour at 420 pounds per square inch, the corresponding flow rate of the gas at 300 pounds per square inch would be 3,500 cubic feet per hour. In tests made at a lower capacity of 3,500 to 4,400 cubic feet per hour, the methane content ran between 8 and 11 percent. This compares favorably with the 7.9 percent made in test 94b at 420 pounds per square inch. In tests 91, 91a, and 91b, operating at a relatively low rate, increased operating pressure did not appreciably increase the methane in the gas. Conversely, to make a synthesis gas for liquid fuels, or for other purposes, a Lurgi generator operating at high rates with a shallow fuel bed would make a low-methane gas. (See fig. 13.)

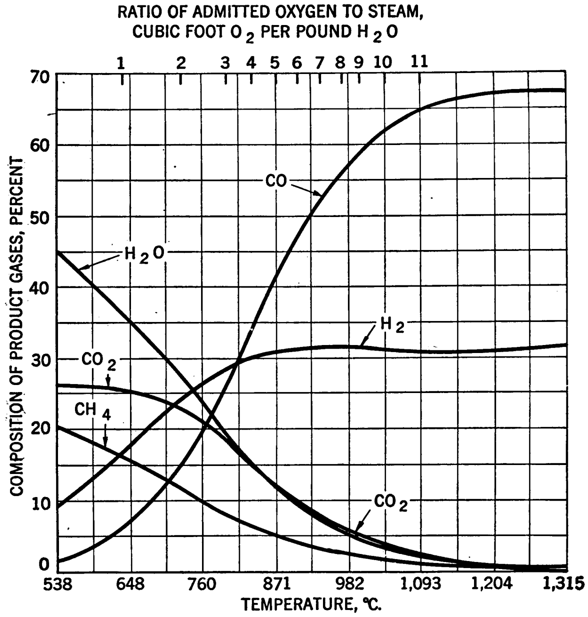

FIGURE 12.—TEMPERATURE DEPENDENCE OF EQUILIBRIUM COMPOSITIONS OF CARBON-WATER-OXYGEN SYSTEMS.

Tests 94c, 94d, and 94e were made at 300 pounds per square inch pressure, but part of the steam in the steam-oxygen mixture was replaced by CO_2. The results of these tests indicate that by introducing CO_2 in the generator the H_2 : CO ratio can be varied over a wide range—from 2.5 down to 0.8—without any difficulty. Synthesis gas for any purpose can be made in this manner. In commercial

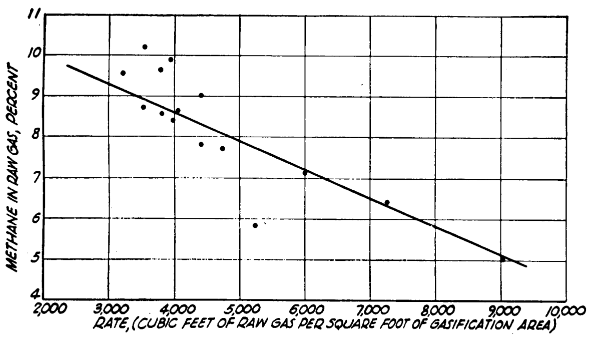

FIGURE 13.—METHANE CONTENT OF RAW GAS VERSUS RATE OF GASIFICATION.

operation the CO_2 washed out of the raw Lurgi gas could thus be utilized.

In test 95 (not shown in table 16), a weathered sample of Hiawatha coal from Utah was tested in the batch generator. Although this coal is classified as bituminous, it has such poor coking qualities that it was thought it might be gasified without previous treatment. It had been demonstrated in tests 76 and 77 that the unweathered coal coked in the generator and could not be used without treatment in the Lurgi process. To prepare the weathered charge, a 100-pound sample was crushed to ½- by ¼-inch size and spread out in the open for approximately 4 months. Test 95 was started in the usual manner, but it soon became apparent that the charge was sticking. When the generator was opened it was found that the coke was very similar to that made in the test with the unweathered sample. In table 16 it will be noted that three tests (88, 89A, and 89B) were made with Hiawatha char. Unlike the chars made from good coking coals, this char was exceedingly easy to make. A 1,200-pound charge without addition of char breeze was placed in the carbonizer and heated as quickly as possible to 350° C. and the heat then turned off. However, unlike the other coals, this coal does not ball up in the retort, so that all of the coal fines in the original charge come out as char fines.

To round out the present program, the last two tests (96 and 97) were made with Pittsburgh-seam, high-temperature coke obtained from the Neville Co., Pittsburgh, Pa. Since all reactivity tests indicate that high-temperature coke is a relatively unreactive material, it was not surprising that the oxygen consumption was higher than in tests made with char. In the zone of methane formation, in the upper part of the generator, the poor reactivity resulted in a gas low in methane. The average methane content of the gas made with the coke is about 2.3 percent as compared to 7 to 9 percent in the gas made with char in tests operating at comparable capacities.

COMPARISON OF VARIOUS GASIFICATION PROCESSES

Since the first phase of the program is completed, it might be advantageous in planning future programs to make some comparisons between the results obtained in the experimental generator and those obtained with various other commercial processes. Of course, such comparisons would be rough. For example, in comparing a unit 13½ inches in diameter with commercial units approximately 9 feet in diameter, the results are influenced not only by the inherent differences in the processes but by the differences in the heat losses and in the type and size of the gasifying materials. The results obtained with commercial units are for long-period operation as compared with 20- to 27-hour runs in the test unit. If the proper prospective is maintained, however, there is some value in the comparison tabulated in table 17. Information on the operation in Germany was obtained by the American team that visited the German plants under the auspices of the Combined Technical Oil Mission of the British and the United States Governments and that reported its findings in various publications.[18] [19] [20] Table 17 summarizes the data for the water-gas and three German processes as given in Odell's publication.[21] The experimental Lurgi test 92, data for which are summarized in table 16, was used for comparison in table 17. This test was chosen because it has a capacity that could be obtained with any char tried in this series of tests. The steam boiler had to be operated at

[18] See footnotes 4 and 5.
[19] See footnote 6.
[20] See footnote 7.
[21] See footnote 6.

TABLE 17.—*Operating data for different gasification processes*

Generator	Standard water-gas process [1]	Lurgi (20 atm. pressure) water-gas process [1]	Winkler continuous water-gas process [1]	"Slagging" producer-gas process [1]	Experimental Lurgi pilot generator [2]
Gas analysis, percent:					
H_2S	----------	1.6	1.2	----------	0.2
CO_2	5.5	32.1	23.1	3.5	29.2
Illuminants	.2	.7	----------	----------	.2
O_2	----------	.2	----------	----------	.2
CO	40.0	12.1	29.1	68.0	19.8
H_2	50.0	37.5	44.2	28.0	41.4
$CH_4+C_2H_6$	.8	14.5	.8	----------	7.1
N_2	3.5	1.3	1.6	.5	1.9
B. t. u./cu. ft. (dry gas, 60° F. and 30″ Hg)	303	333	253	311	281
B. t. u/cu. ft. (CO_2- and H_2S-free)	321	502	334	322	398
Fuel, kind	H. T. coke	Partly dried brown coal	Carbonized brown coal	H. T. coke	Low-temperature char
Size, inches	----------	0.12 by 0.40	0.12 by 0.40	1.5 and over	0.125 by 0.75
B. t. u./pound	----------	8,135	10,190	[3] 12,500	12,990
O_2/M cu. ft. raw gas (60° F. and 30″ Hg)	----------	106.6	243.0	250.0	127.0
Steam, lb./M cu. ft. raw gas (60° F. and 30″ Hg)	----------	63.2	29.1	22.4	78.8
Fuel, lb./M cu. ft. raw gas (60° F. and 30″ Hg)	----------	67.8	47.04	26.7	24.36
Fuel/M cu. ft., CO_2- and H_2S-free (60° F. and 30″ Hg)	----------	102.0	62.15	27.7	34.5
Raw gas/ton fuel, cu. ft. (60° F. and 30″ Hg)	----------	29,500	42,500	74,900	82,100
$\text{Eff.}=\frac{\text{B. t. u. in gas}}{\text{B. t. u. in fuel}}$	----------	[4] .60	[4] .53	[4] .93	[4] .89
Capacity, cu. ft./sq. ft. per hr	2,290	1,990	[5] 10,500	6,350	7,246
Therms/sq. ft	6.94	6.3	24.96	19.74	20.36

[1] Values taken from W. W. Odell, Bureau of Mines, Inf. Circ. 7415, table 4.
[2] Test 92, table 16.
[3] B. t. u. estimated from coke analysis.
[4] Calculated.
[5] This figure probably is too high; see reference 5.

its full capacity, and the gas stream had to be divided and measured with two meters; otherwise, the operation and the results obtained were similar to those in other tests. There were no indications that the generator producing 7,246 cubic feet of gas per hour corrected to 60° F. and 30 inches Hg or operating 9,000 cubic feet per hour for the last hour had reached its capacity. Moreover, there were some indications that the operation was proceeding more smoothly at these high capacities. For example, the combustion zone—that is, the demarcation between ash and unburned carbon—was more clearly defined.

In table 17, it is indicated that the Lurgi generators require appreciably less oxygen than the other two types, the requirement for the commercial Lurgi being slightly less than for the experimental unit. The thermal conversion efficiency, $\frac{\text{B. t. u. in gas}}{\text{B. t. u. in fuel}}$, is highest for the slagging-type generator, slightly less for the experimental Lurgi, still lower for the commercial Lurgi, and least for the Winkler. Probably the wide difference in efficiencies of the commercial and experimental Lurgi units is due largely to the superior grade of fuel used in the latter. No comparison can be made on gasification of the various ranks of coals in the experimental Lurgi unit, since only one uncompleted anthracite test was made and none with lignite and subbituminous coals.

OUTLINE OF TESTS REQUIRED FOR PURIFICATION AND SYNTHESIS

Little additional work has been done on purification of the gas since installation of the continuous generator. The few trials made confirm the previous findings that the soda-iron oxide catalyst is a slow-acting material. The best results were at a space velocity of 200; the organic sulfur was reduced from about 10 grains (in gas from Corona-seam coal) to 0.30 grain per 100 cubic feet of gas. If subsequent testing substantiates these results, the question will arise whether it is economically feasible to use this catalyst in commercial operation, since the size of the apparatus to purify the gas would be 10 to 15 times the size required to synthesize the gas. To purify the gas still further, an activated-carbon vessel was put in the train after the soda-iron oxide purifiers. Laboratory tests with carbon showed that sulfur concentrations as low as 0.01 grain per 100 cubic feet of gas were obtained. However, no attempt was made to measure the life of the carbon; and until this is done, very little information is available for evaluating all economic factors involved in using this material. The Morgantown, W. Va., Station of the Bureau of Mines is engaged in an elaborate test program of gas purification. It plans to test standard methods of purification and devise some new methods. Its findings may be the basis for future work done in purifying Lurgi gas.

Long tests to determine the life of the catalyst have not yet been made for two reasons. First, the problems involved in reducing the sulfur in the gas to values low enough to make life tests on catalysts have not yet been solved satisfactorily in the test operation. Indications are that the catalyst is poisoned in less than 24 hours in the present apparatus, using gases containing 0.3 grain of sulfur per 100 cubic feet. In addition, the capacity of the converter is about five times that of the purification train, so that little information other than the amount of sulfur absorbed by the catalyst before it is inactivated would be obtained if this relatively small amount of gas were synthesized per unit of time. In the second place, to make gas for 6 months continuously in the generator would require enlarging the present carbonization capacity. The amount of char gasified in low-capacity tests was about 100 pounds per hour or 2,400 pounds per day. Since the maximum capacity of the carbonizer is about 800 pounds of char per charge, 3 days would be required to make enough char for 1 day's operation of the gasifier. Since successful operation of the Lurgi generator, as demonstrated by the work reported here, depends on using bituminous-coal char, the whole question of low-temperature carbonization will have to be reexamined very thoroughly; for this reason, it would be unwise at present to either enlarge or build a new batch carbonizer.

APPENDIX

Details of a typical test will be described here. Test 86, which is typical of those made in the pilot plant, was started at 8:00 a. m., April 28, 1948, by slowly heating the superheater, through which air was flowing at a rate of 400 cubic feet per hour, to a temperature in the neighborhood of 150° C. The hot air also dried and heated the ash over the grate. At 9:30 a. m. a pail of red-hot char was thrown into the generator through the charging hopper. When thermocouple 4, which measures the liner temperature at the gasification zone, indicated that the hot char was burning, a pail of cold char was added. Additional char was added intermittently until 10:15 a. m., when the generator was full. The top valves were closed, and the gas was passed through the cooling train and the meter. As the liner temperature approached 500° C., the air was progressively saturated at higher temperatures with steam. At 10:50 a. m. the saturated air was replaced with the oxygen-steam mixture. The pressure regulator was set to increase the pressure in the generator. At 12:00 noon the operating pressure of 300 pounds per square inch was reached. The gasification rate was approximately 1,100 cubic feet per hour and was slowly increased to about 4,400 cubic feet per hour by 4:00 p. m. The values reported for the test in table 16 are those from 2:30 p. m. to 8:00 a. m. on April 29, when the steam and oxygen were turned off.

Charts from test 86 are shown in figures 14, 15, 16, 17, and 18. The CO_2 (fig. 14) and the specific-gravity (fig. 15) recorders are direct-reading instruments. The specific-gravity chart covers a portion of the test. The apparent increase of the specific gravity of the gas is a result of the chart being put in slightly off center. The oxygen (fig. 16) and steam (fig. 17) flow rates were recorded on logarithmic scales, which were calibrated in cubic feet of oxygen and pounds of steam per hour, respectively. The temperature chart (fig. 18) covers about 2.5 hours of the test run. The millivolt readings shown are from chromel-alumel thermocouples placed in the gas stream (*1*) before the superheater, (*2*) after the superheater, and (*3*)—which was not recording accurately in this test—at the grate. The reading for *3* is usually 0.5 millivolt under *2*. Thermocouples *4*, *5*, and *6*, give the readings of the liner temperatures. The bunching of these points indicates that the high-temperature zone was below the *4* thermocouple. Ordinarily, if the gasification zone got too close to the grate, *3* would read above *2*. Thermocouple *7* records the temperature of the effluent gas at the generator, while *8* shows the temperature after the precooler but before the pressure valve. Thermocouples *9*, *10*, and *11* are in the synthesis train, which was not in operation during this test. Thermocouple *12* records the temperature inside of the ash hopper. It will be noted in figure 17 that the steam-flow rate was increased slightly after 4:45 p. m. In figure 14, at 4:45 p. m., the CO_2 in the effluent gas was decreasing. Since the ash characteristics of this char were not well-known, it was feared that a gasification temperature corresponding to 26–27 percent CO_2 in the effluent gas would form clinkers. Accordingly, the steam : oxygen ratio was slowly increased until 5:45 p. m., when the CO_2 in the effluent gas started increasing. Generally, small variations in the steam : oxygen ratio and in the superheater temperature immediately affect the CO_2 content of the gas.

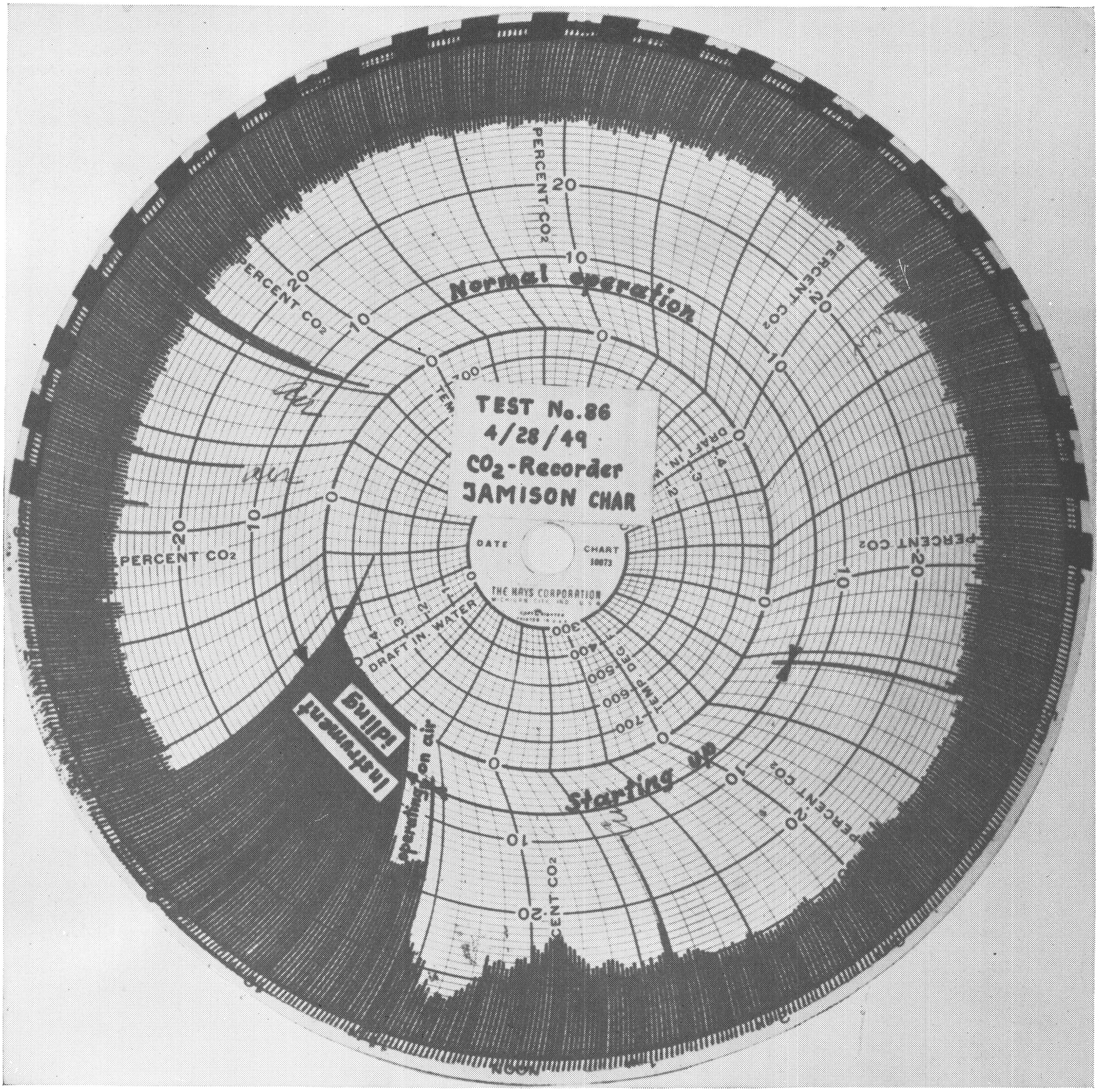

FIGURE 14.—CARBON DIOXIDE CHART.

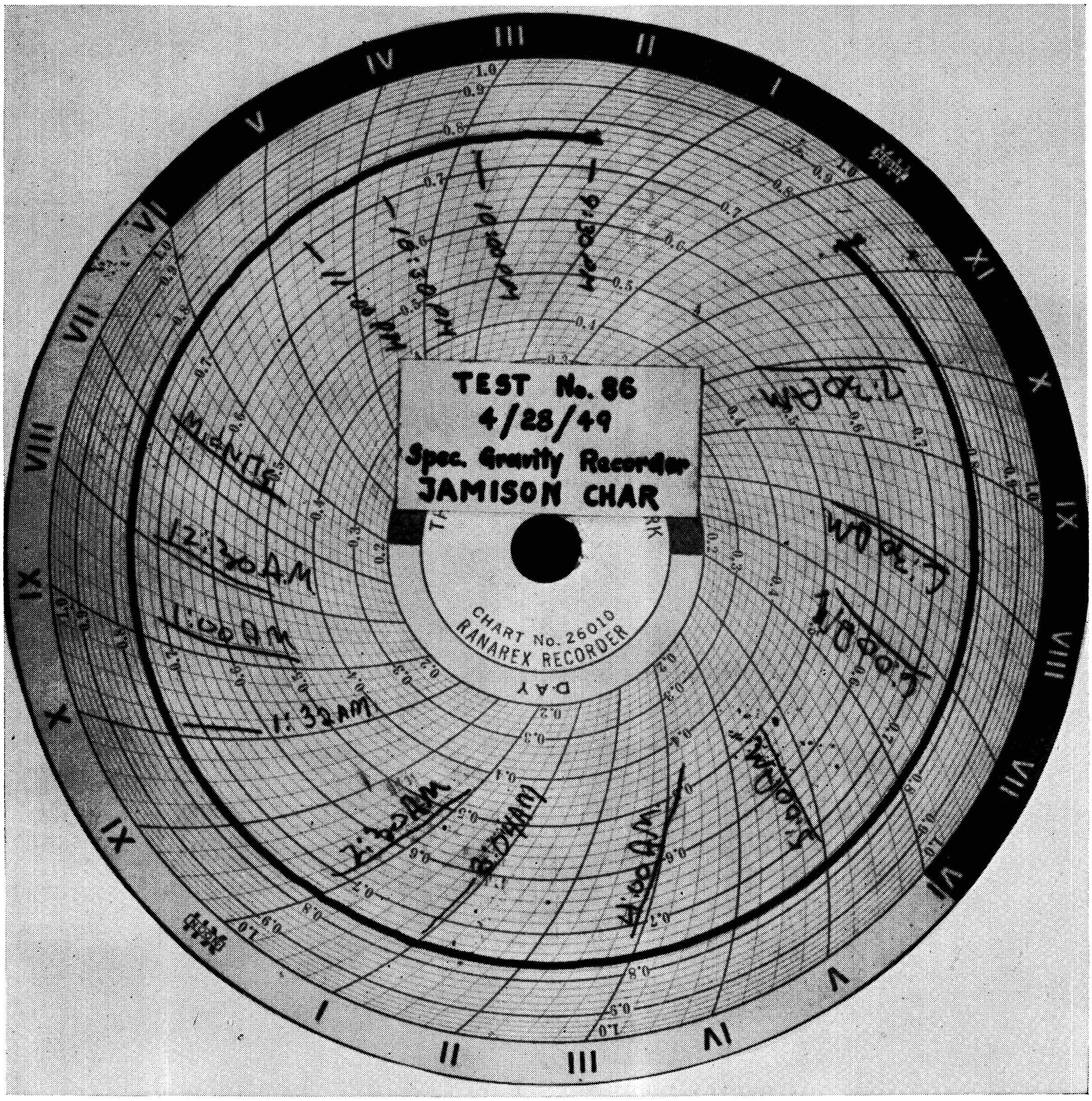

FIGURE 15.—SPECIFIC-GRAVITY CHART.

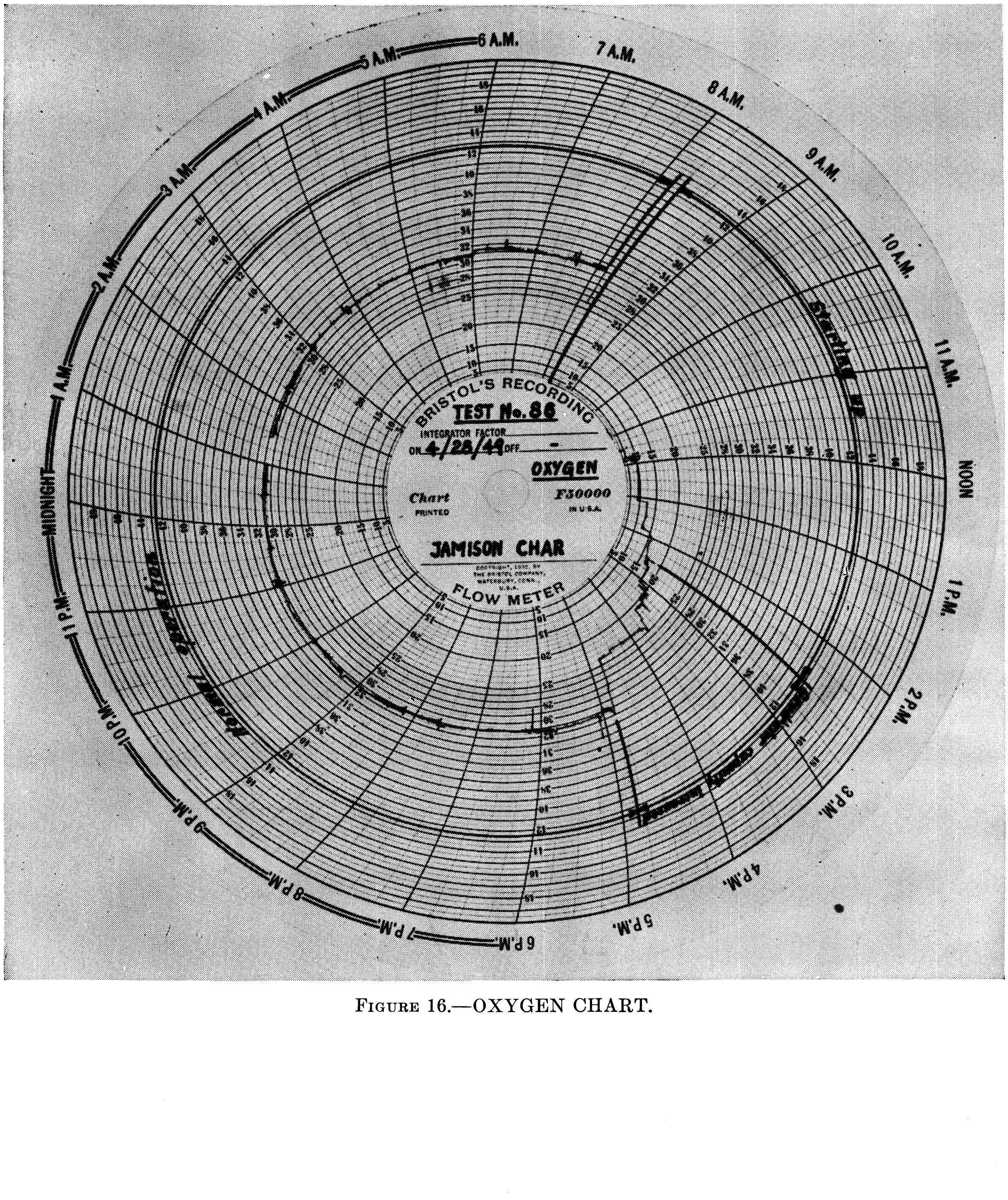

FIGURE 16.—OXYGEN CHART.

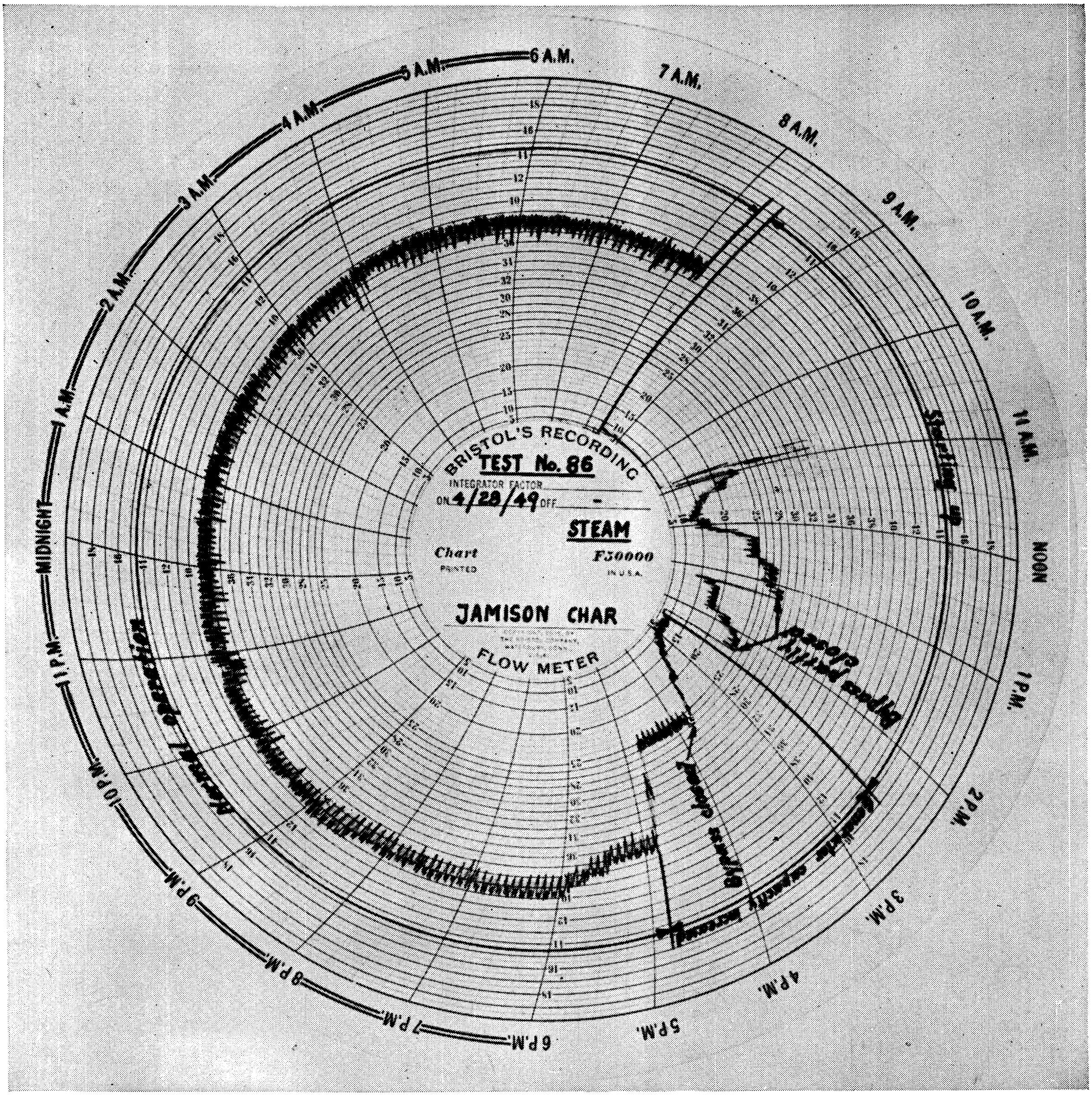

FIGURE 17.—STEAM CHART.

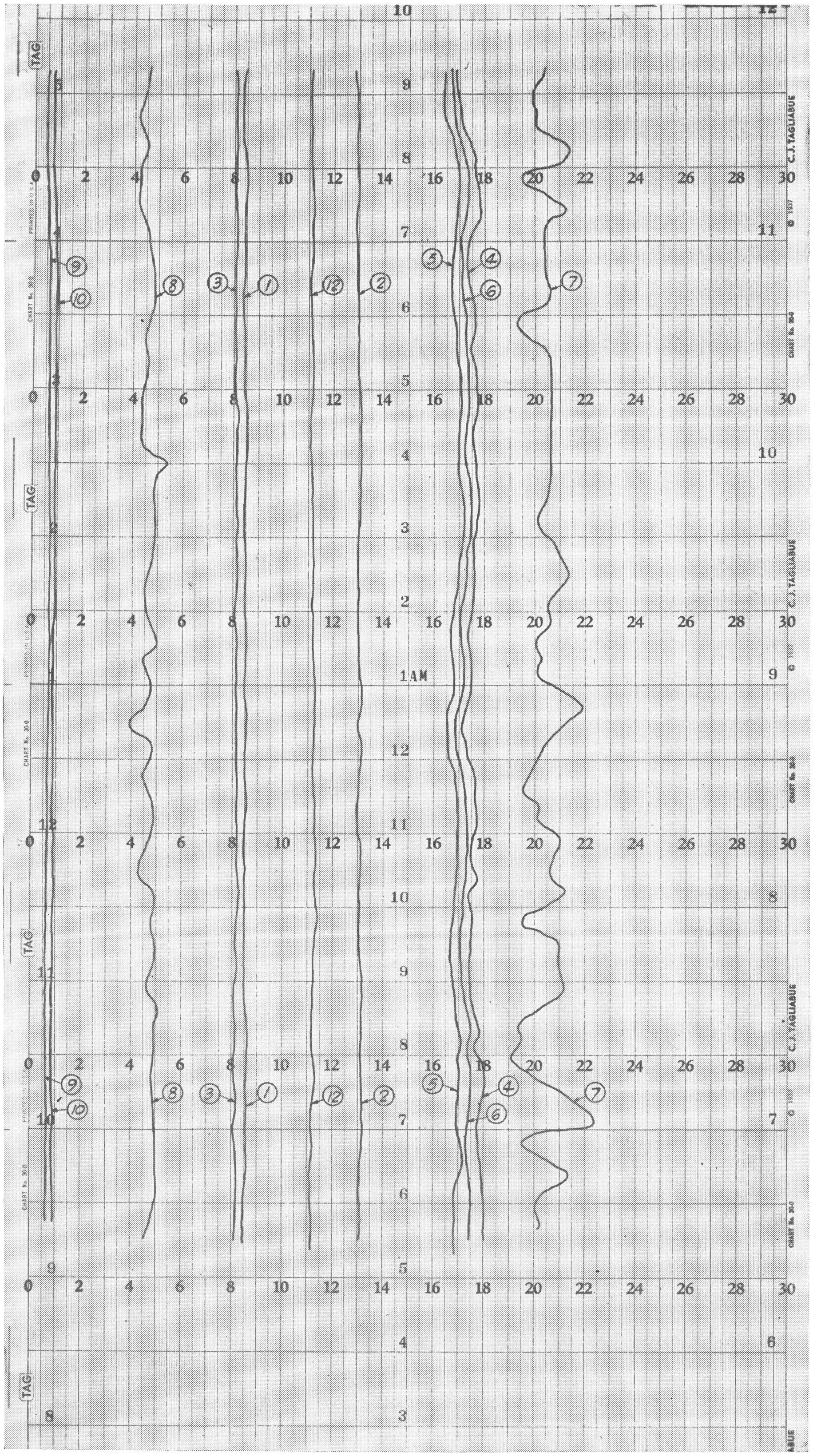

FIGURE 18.—TEMPERATURE CHART.