steam and products of gasification, which produced a thin layer of iron-oxide scale. Appearance of the lower section of the inner tube after run 16, when it had been in use for nearly 3,800 hours, is shown in figure 21, showing formation of scale blisters. Although the use of mild steel for the inner tube did not cause operational difficulties, a heat-resistant alloy should be used for a commercial plant.

SECTION II. SUMMARY OF WORK ON GASIFICATION PROJECT PREVIOUS TO JULY 1, 1950

The problem of producing high hydrogen gas from lignite for partial or total reduction of taconite was assigned to the Bureau of Mines in August 1943 (12). Previous experience had shown that natural lignite could not be satisfactorily gasified in a conventional water-gas generator owing to excessive degradation. In late 1943 an attempt to gasify lignite-char briquets in a conventional water-gas set was unsuccessful because of disintegration and low efficiency caused by loss of fine carbon (12).

Evaluation of various gasification processes in relation to known physical and chemical properties of lignite indicated that the Reyerson-Gernes process, developed on a laboratory scale at the University of Minnesota, might be used for continuous gasification of lignite in an externally heated reactor. The reactor consisted of two concentric steel cylinders separated by a 3-inch annular space. The outer cylinder was heated by a surrounding cylindrical furnace. Lignite fed at the top of the reactor slowly descended by gravity through the annular space where gasification occurred by reaction of carbon of the lignite with steam admitted simultaneously with the lignite. As a result of high chemical reactivity of lignite, rate of formation of water gas was found to be quite high without exceeding maximum temperatures allowable in a steel-tube reactor.

A retort and heating system designed by V. F. Parry was chosen to investigate the engineering possibilities of the process. Construction of two units of different capacity was planned; one, a small pilot plant was constructed at existing Bureau facilities at Golden, Colo., for preliminary experimental testing, and the second, a larger, commercial-size unit, was established at Grand Forks, N. Dak.

The small pilot plant having a design capacity of 2,500 cubic feet of water-gas per hour was completed at Golden in February 1944. The original unit consisted of 2 concentric, standard-steel pipes 12 and 6 inches in diameter, respectively, forming a 3-inch wide annular reaction space. Heat for the endothermic gasification reactions was transferred through the outer metal wall from a surrounding combustion chamber.

This pilot plant was operated in 1944 for approximately 1,000 hours. During this period, 1.5 million cubic feet of gas was produced from 24 tons of natural lignite, steam-dried lignite, and subbituminous coal. Gas with hydrogen-carbon monoxide ratios ranging from 1.6 to 12.0 was made at rates up to 65 cubic feet per square foot of heated tube surface per hour.

A mild-steel reaction tube used initially in the small pilot plant unit failed by oxidation and carburization after about 550 hours of operation. A steel pipe processed on both the inside and the outside by a sprayed metallic coating of chromium-nickel alloy was used without serious deterioration for the remainder of the 1,000-hour testing period. Detailed information and design and operation of this plant was reported by V. F. Parry and others (12).

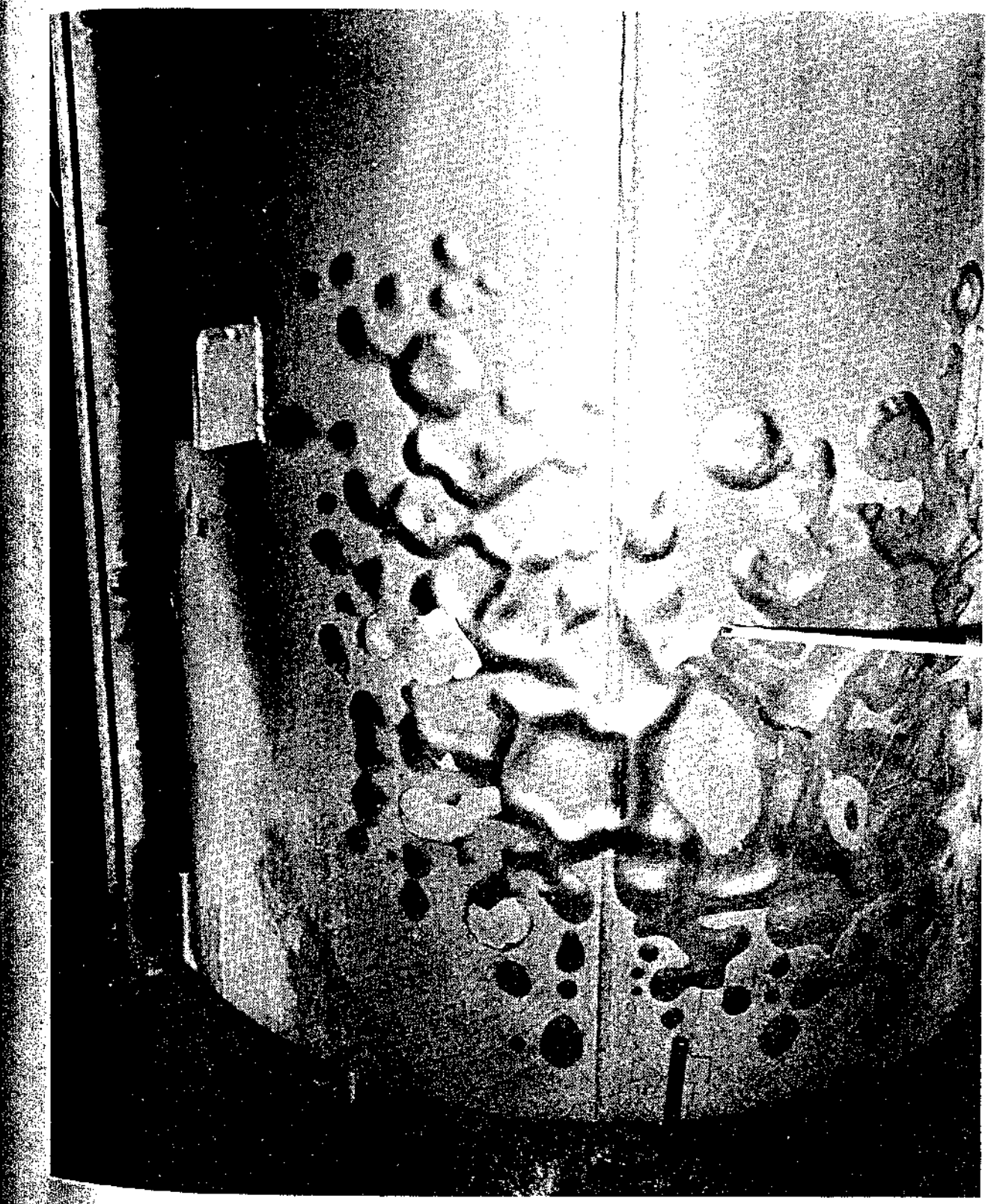

Figure 21. - Lower section of mild-steel inner tube after run 16.

Subsequently, the small pilot-plant gasifier was modified by increasing the diameter of the externally heated reaction tube from 12 inches to 16 inches and reducing the width of the reaction space from 3 inches to 2 inches. The plant operated smoothly at gas rates as high as 3,300 cubic feet per hour while processing subbituminous coal and lignite. Conversion of carbon of the coals to gas ranged from 64.2 to 78.6 percent, depending on feed rate and rate of steam admission. A low ratio gas having a hydrogen-carbon monoxide ratio of from 1.92-2.11 was produced during this second series of tests (13).

Construction of the commercial-scale pilot plant was completed at Grand Forks, N. Dak., early in 1945. Details of construction and information obtained during 1,800 hours of operation were reported by V. F. Parry and others (13). The large pilot plant embodied the essential features of the small unit scaled up on a production basis by a factor of about 6 to 1. The outer reaction tube was approximately 4 feet in diameter and 20 feet in length.

In initial tests of the large gasifier, a spray-coated reaction tube similar to that tested in the small plant was used. After some 450 hours of operation, this tube was replaced by an alloy-clad tube consisting of a 3/8-inch, mild-steel inner core between integral 1/8-inch-thick layers of 26-percent chromium steel.

The first series of tests showed that the large gasifier was technically feasible and that product gas of varied composition could be made readily. Most major operational difficulties were eliminated, and the relative influence of many variables was determined. It was shown that the composition of the product gas could be readily altered over a wide range by changing steam concentration and temperatures in the combustion space. Hydrogen-carbon monoxide ratios ranging from 2.15 to 5.43 were obtained in the first 4 experimental runs. Between 47 and 85 percent of the carbon in the feed was gasified.

Results of an additional 5 runs totaling about 2,000 hours of operation from late 1946 to December 1948 were reported by A. C. Burr and others (4). In addition to the development of the technology of lignite gasification and investigation of process variables, main attention was given to performance of the externally heated reaction tubes and to proving out a satisfactory tube life under gasification process operating conditions. The double-armored reaction tube corroded excessively in some 1,780 hours of operation, of which nearly 40 percent was at combustion-space temperatures over 2,000° F. After reconditioning, the tube was used for an additional 1,530 hours at combustion-space temperatures under 2,000° F. Corrosion was reduced but progressive deformation, apparently caused by difference in thermal expansion between the chromium alloy cladding and the mild-steel core, occurred to such an extent the tube was abandoned.

A third reaction tube, constructed from two centrifugally cast sections of chromium-nickel steel, was then installed. A test run with this tube and with width of the annular reaction space reduced from 3 to 2-1/4 inches was terminated after only 98 hours of erratic operation. As a result of the rough inner surface of the cast reaction tube and the reduced width of the annulus, the lignite hung up at various locations in the reaction space, causing unsatisfactory operation.

During this series of runs, the previous batch-wise method of feeding lignite to the gasification retort was changed by installing a reciprocating feeder and rotary block valve so that lignite could be added to the charging dome in small 2.0- to 3.5-pound increments. This method of feeding prevented disturbance in the thermal equilibrium of the reactor while lignite was being added.

Hydrogen-carbon monoxide ratio of the gas generated was varied from 2.3 to 9.0. Rate of gas production was as high as 11.9 M cu. ft. per hour, corresponding to 60 cu. ft. per sq. ft. of heated surface per hour, and gas yields were as high as 91 M cu. ft. per ton of moisture- and ash-free lignite.

The highest hydrogen-carbon monoxide ratios of the product gas were obtained by greatly increasing the rate of steam admission. Concentration of hydrogen in the product gas changed relatively little (from 55.8 to 61.1 percent) when the hydrogen-carbon monoxide ratio changed from 2.3 to 9.0. However, percentage of carbon monoxide was reduced from 24.3 to 6.8 percent and that of carbon dioxide increased from 13.4 to 26.8 percent. In general, experience showed that production of a low ratio gas is more desirable from the standpoint of ease of operation and gas-making capacity of the gasification unit because of the large quantities of unreacted steam passing through the gasifier in operation at the highest H_2-CO ratios. In a test at the highest H_2-CO ratio, 9.0, the volume of steam leaving the gasifier was more than twice the volume of dry gas generated.

Four additional runs totaling 1,915 hours of operation were completed during the period from January 1, 1949, to June 30, 1950 (3). Twenty million cubic feet of gas was produced from 428 tons of steam-dried and natural lignite. Gas-production rates were from 7.8 to 16.4 M cu. ft. per hour, the latter being the highest yet reached. Hydrogen to carbon monoxide ratios ranged from 1.85 to 6.5. Both the highest gas-production rate and the lowest hydrogen-carbon monoxide ratio were obtained when processing steam-dried lignite.

Performance of the alloy-steel reaction tubes received considerable attention. The cast-alloy tube failed when it cracked at part of the circumferential weld seam joining the two cast sections. No attempts were made to recondition this tube. On the basis of previous experience, a retort tube fabricated from 310-alloy rolled plate was selected for testing. In nearly 1,900 hours of operation to June 30, 1950, this tube was found to resist corrosion satisfactorily, but some deformation was noticeable, which, however, was not enough to cause operational difficulty.

Results of experiments to determine the best location for admitting process steam when operating a divided annulus arrangement showed that under otherwise comparable experimental conditions more carbon was gasified when at least a portion of the process steam was admitted to the upper reaction zone. Increase in the rate of steam addition to the upper reaction zone had a greater effect on carbon gasified and gas made than an equal increase in rate of steam addition to the lower reaction zone.

In tests of operation at various lignite feed rates, results indicated that percentage of carbon gasified, and, consequently, the gas yield per ton of lignite decreased with increasing feed rate, whereas hydrogen-carbon monoxide ratio of the product gas increased. This was chiefly the result of decreasing effective length of the reaction zone available for gasification because an increasingly longer zone was needed to dry and preheat the lignite to reaction temperature with an increased feed rate. Because percent carbon gasified did not decrease in proportion to increase in lignite feed rate, the total gas generated increased as lignite feed rate was increased. At the highest lignite feed rate and lowest carbon gasification (53 percent), tar carryover increased sharply, causing operating difficulties in the retort and gas-scrubbing system.

Some 250 tons of lignite dried by the Fleissner process was also gasified under conditions closely comparable to tests run on natural lignite. At equal lignite feed

rates on moisture- and ash-free basis, carbon gasified and gas made were substantially higher when dried lignite was gasified because of the reduced length of reaction space required for drying and the longer zone available for gasification. At the higher combustion-space temperatures, and for equal ratios of total water available to moisture- and ash-free lignite, the increase in gas-making capacity using dried lignite was as much as 30 percent.

SECTION III. CORRELATING METHODS FOR OVERALL GASIFICATION DATA

In presenting gasification results for individual runs, a number of correlating plots were developed to show the influence of individual process variables over a relatively narrow range of operating conditions. In general, as previously discussed, these correlating plots present the results of groups of experiments in which an attempt was made to hold most operating conditions constant and evaluate the individual effect of a single process variable. The results of a few additional correlating methods of more general nature that bring out certain relationships among the annular-retort gasification data as a whole are presented in this section. These plots also show in summary form the range and extent of the rate of gas production, gas composition, and other variables for the several gasification runs.

Gas Composition in Terms of Water-Gas Equilibrium

The analysis and correlation of gasification data in terms of equilibrium of the water-gas shift reaction has been proposed by a number of authors. Some experimental evidence is available that indicates the probable approach to equilibrium of this reaction in various gasification systems (8, 10, 11). Batchelder and Sternberg have developed a method for thermodynamic analysis of gasification systems (2), based on the assumption of water-gas shift equilibrium at the exit temperature from the gasifier. Batchelder and Sternberg have developed and applied their analysis primarily for the oxygen-steam gasification of pulverized coal. Application of the method has been elaborated by Edmister, Perry, Corey, and Elliott (9), who present a number of charts that are designed to facilitate the solution of the material balance and enthalpy relationships. The resulting correlations show the predicted relationships of composition of the product gas to ratios of the feed materials and to gasifier heat losses. In addition to making it possible to predict the probable effect of variation in the ratios of the feed streams, the correlations permit the evaluation of materials requirements per unit of product gas under various operating conditions.

The assumption of water-gas equilibrium at all points and temperature levels has also been used by Batchelder and Busche in their detailed theoretical study of gasification rate in the gasification of pulverized coal with oxygen and steam (1).

Data from the annular-retort gasifier have been obtained for a wide range of operating conditions, and in particular for a wide range of steam-lignite ratios that might be expected to alter gas composition by water-gas shift. Preliminary correlations of these gasification data (5) have indicated the probability that composition of the product gas was controlled by water-gas shift. In figure 22, experimental data for composition of product gas from the annular-retort gasifier are plotted versus steam-lignite ratio and are compared with calculated curves based on water-gas shift equilibrium.

The experimental data plotted in figure 22 include all test periods for gasification of natural lignite from runs 3 through 13 and cover the maximum range of steam-lignite ratios that were investigated. Additional data from runs 14 through

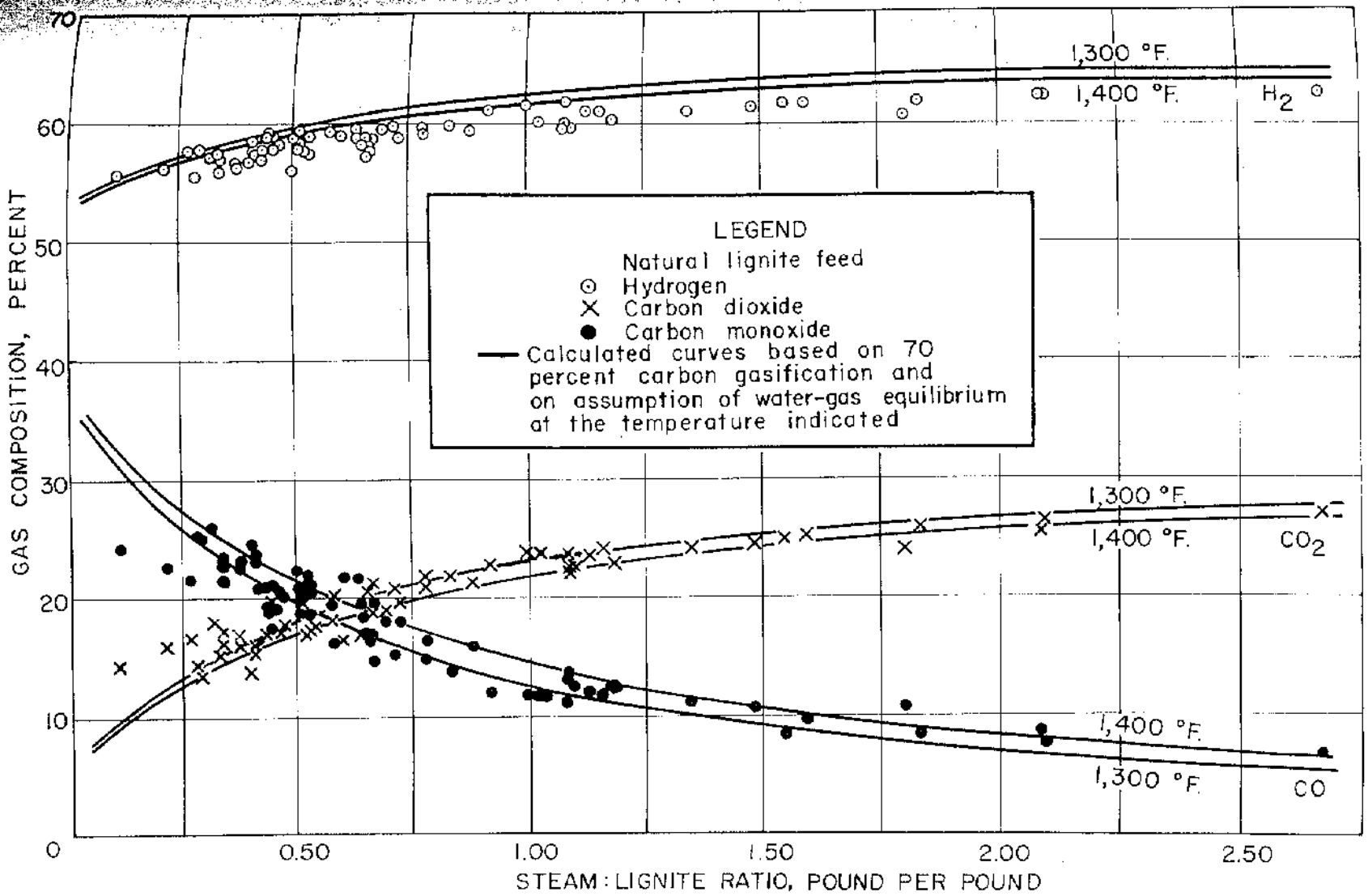

Figure 22. - Gas composition as a function of steam-lignite ratio; comparison with calculated curves based on water-gas equilibrium.

17 are found to follow the same trend but have not been included because they fall mainly within a relatively narrow range of steam-lignite ratios.

The calculated curves in figure 22 are based on the assumption of gasification of 70 percent of the carbon in the lignite as charged. Most of the experimental data were obtained within a range of 60 to 80 percent carbon gasified. Two curves are given based on the assumption of water-gas shift equilibrium in the product gas at 1,300° and 1,400° F.

Typical analyses of the natural and steam-dried lignite that were used in determining the calculated curves are given in table 15.

TABLE 15. - Typical lignite analyses used in calculation of equilibrium composition of product gas

Ultimate analysis, percent	Natural lignite	Dried lignite
Oxygen	44.42	28.61
Hydrogen	6.83	5.19
Sulfur	.70	1.25
Nitrogen	.55	.75
Carbon	41.50	57.10
Ash	6.00	7.10

In the calculation the following assumptions were made with respect to minor components: (1) Small amounts of methane and other hydrocarbons in the product gas were considered only for material balances. From experimental data, the average amount of methane and heavier hydrocarbons appearing in the product gas was taken as 4.00 pound mols per ton of natural lignite and 5.32 pound mols per ton of dried lignite as charged (2) Sulfur in the lignite was assumed to combine with equivalent hydrogen to form hydrogen sulfide. (3) Nitrogen was considered as an inert.

In figure 23 the same data are replotted to show the variation in H_2-CO ratio of the product gas with steam-lignite ratio, and the experimental data are again compared with calculated curves based on water-gas shift equilibrium. Included in this figure are results for gasification of steam-dried lignite in run 12. The calculated curve for the dried-lignite gasification data is based on an average carbon gasification of 75 percent and a water-gas shift equilibrium temperature of 1,400° F.

In general, as shown in figures 22 and 23, the experimental data for gas composition are in very good agreement with the calculated curves. This correlation confirms the fact that water-gas shift equilibrium controls the composition of gas obtained from the annular-retort gasifier and indicates the relatively minor influence of other operating factors on gas composition. The assumed temperatures of water-gas equilibrium, 1,300° and 1,400° F., were selected as giving the best representation of the overall data. It is evident that gas composition does not change rapidly with the assumed equilibrium temperature. The assumed equilibrium temperatures that give the best correlation of the data are somewhat higher than the measured temperatures at the gas offtake from the annular retort, which normally fell in the range from 1,100° to 1,250° F.

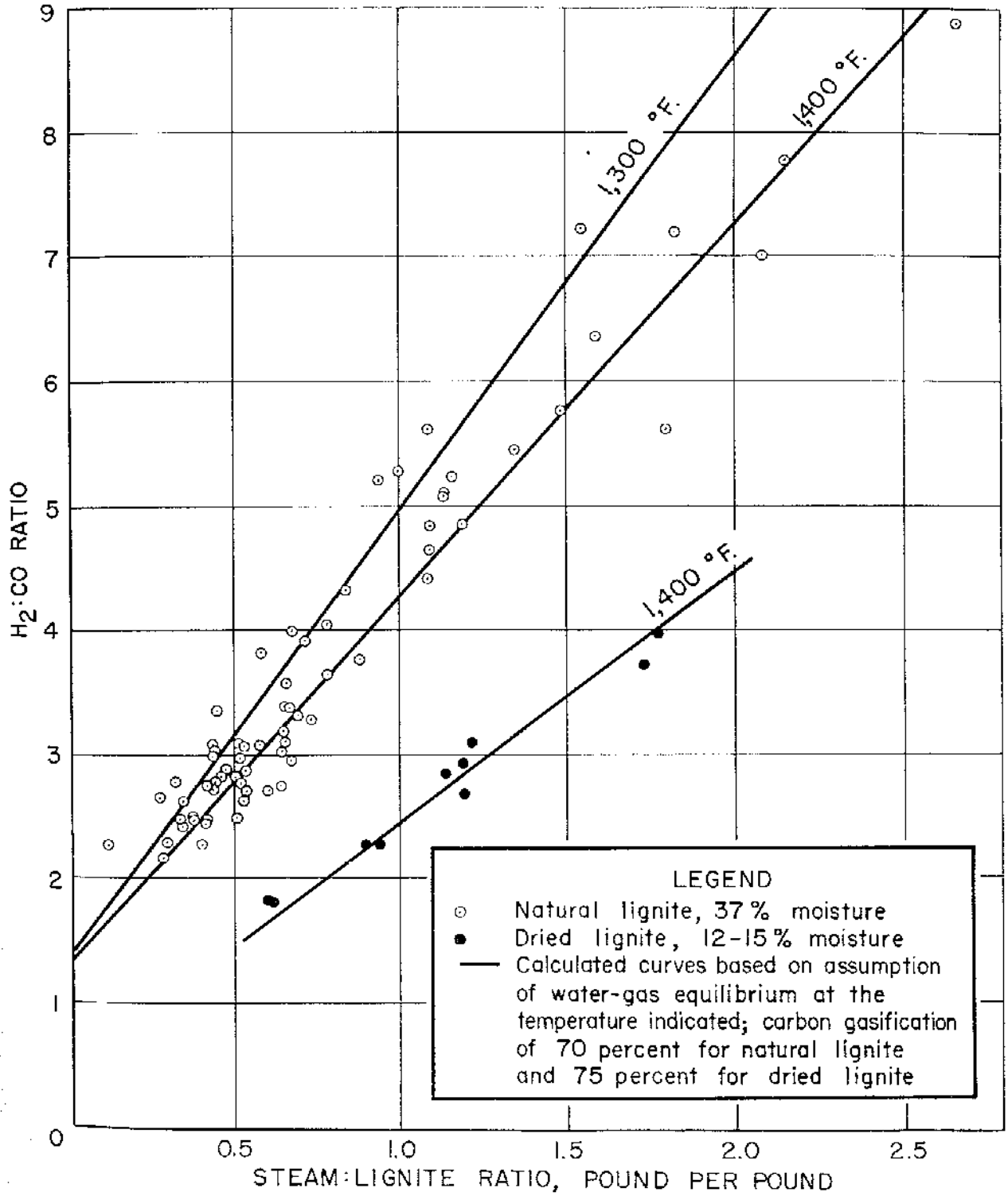

Figure 23. - H_2-CO ratio of product gas as a function of steam-lignite ratio; comparison with calculated curves based on water-gas equilibrium.

Material Balance Relationships

The preceding correlation relates gas composition to the feed ratio of steam to lignite but gives no information on gasification rate. For the gasification of a given lignite in the annular-retort gasifier, it can be shown stoichiometrically that the rate of production of (H_2 + CO), the useful components of synthesis gas, is a function only of the lignite feed rate and the percent carbon gasified.

The amount of (H_2 + CO) is not affected by changes in gas composition and H_2-CO ratio through water-gas shift at a given percent carbon gasification. The relationship between rate of production of (H_2 + CO) and the lignite feed rate is illustrated in figure 24. Experimental data are plotted for gasification of natural lignite in runs 3 through 11 and run 13 and for gasification of steam-dried lignite in run 12. A material-balance line has been placed through the natural-lignite gasification data based on an average carbon gasification of 70 percent. The corresponding line through the dried-lignite gasification data is based on 75 percent carbon gasification. These material-balance calculations were based on the typical analyses of natural and steam-dried lignite that are listed in table 15 and on the same assumptions for effect of minor components that were discussed in the preceding section.

In the group of experiments on gasification of natural lignite, carbon gasified actually was varied from over a maximum range from 54 to 88 percent. In figure 25 the data for (H_2 + CO) produced per pound of lignite, as charged, are plotted versus the percent carbon gasified. A material-balance line based on the typical analysis of table 15 and the various assumptions with regard to minor components is shown on the plot.

The relationships shown in figures 24 and 25 are basically material-balance relationships only and do not provide any information on the process variables such as temperature and steam-lignite ratio that are responsible for effecting a given degree of carbon gasification at a specified lignite feed rate. In general, the relatively complex effect of these process variables on carbon gasification has been separately evaluated only through presentation and intercomparison of more limited groups of data in which a single process variables was varied with others held approximately constant. These correlation plots indicating the effect of individual process variables within a limited range are presented as part of the detailed analysis of gasification data in the present report and in previous progress reports. In general, steam-lignite ratio had a significant effect on percent carbon gasified at otherwise comparable operating conditions, but its main effect, particularly at the higher ratios, was to alter gas composition by water-gas shift. The general relationship of rate of gas generation to heat-release rate and temperature in the gasifier heating furnace is discussed in the following section on thermal requirements.

Thermal Requirements

A preliminary study of the thermal requirements for gasification of lignite in the annular-retort gasifier has been published (6). Heat requirements for this study were defined in terms of total heat released in the external heating furnace surrounding the gasifier, based on net heating value of the gas burned in this external furnace. This study, based on data from runs 3 through 11, indicated that the heat required per standard cubic foot of dry gas generated varied only moderately despite the wide range of operating conditions investigated. Correlations that were developed indicated the thermal requirements for gasification of natural lignite depend primarily on H_2-CO ratio, ranging from 135 to 107 B.t.u. per standard cubic foot of dry gas between the H_2-CO ratios of 2 and 9.

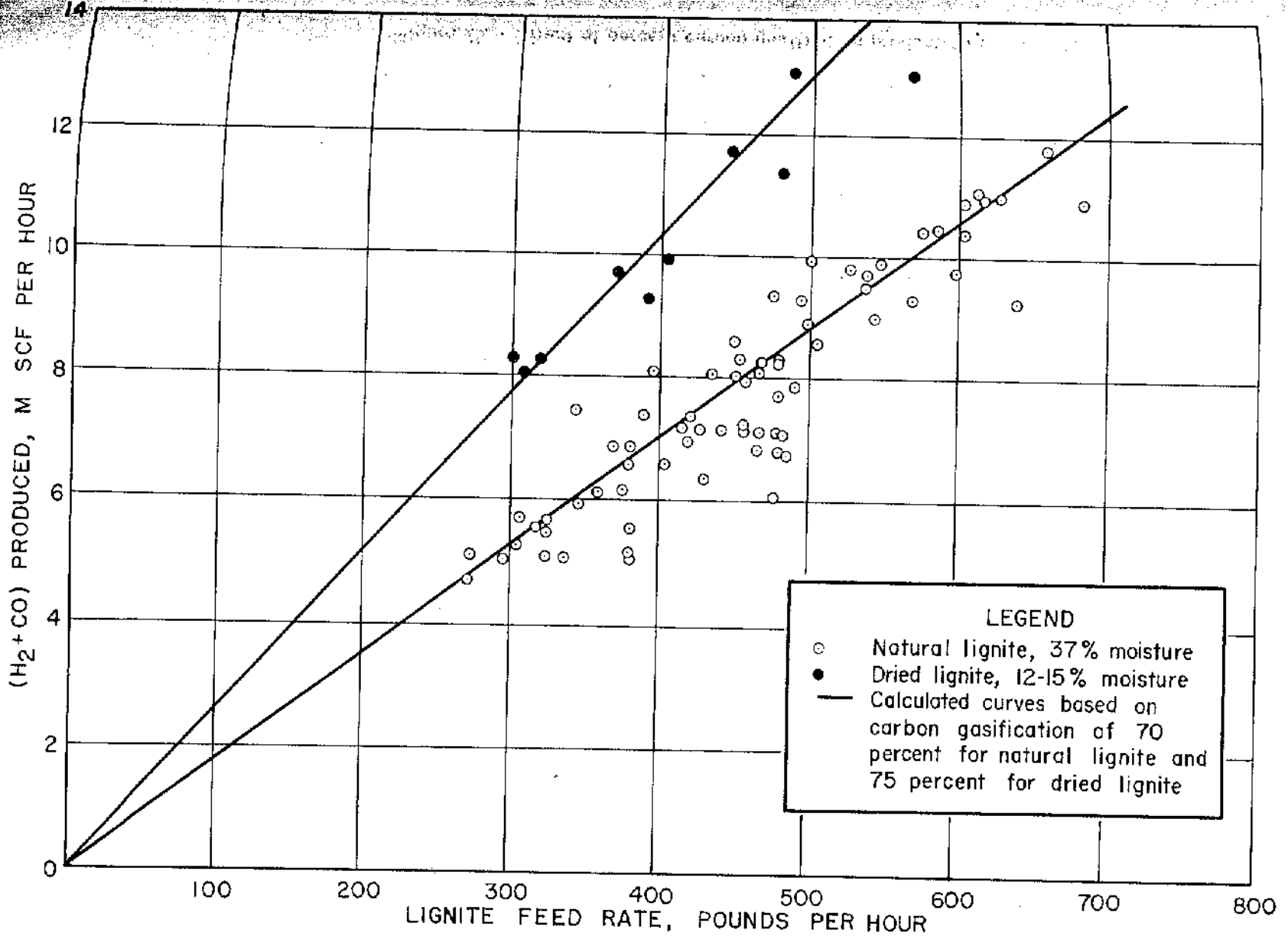

Figure 24. - Rate of production of (H_2+CO) as a function of lignite feed rate.

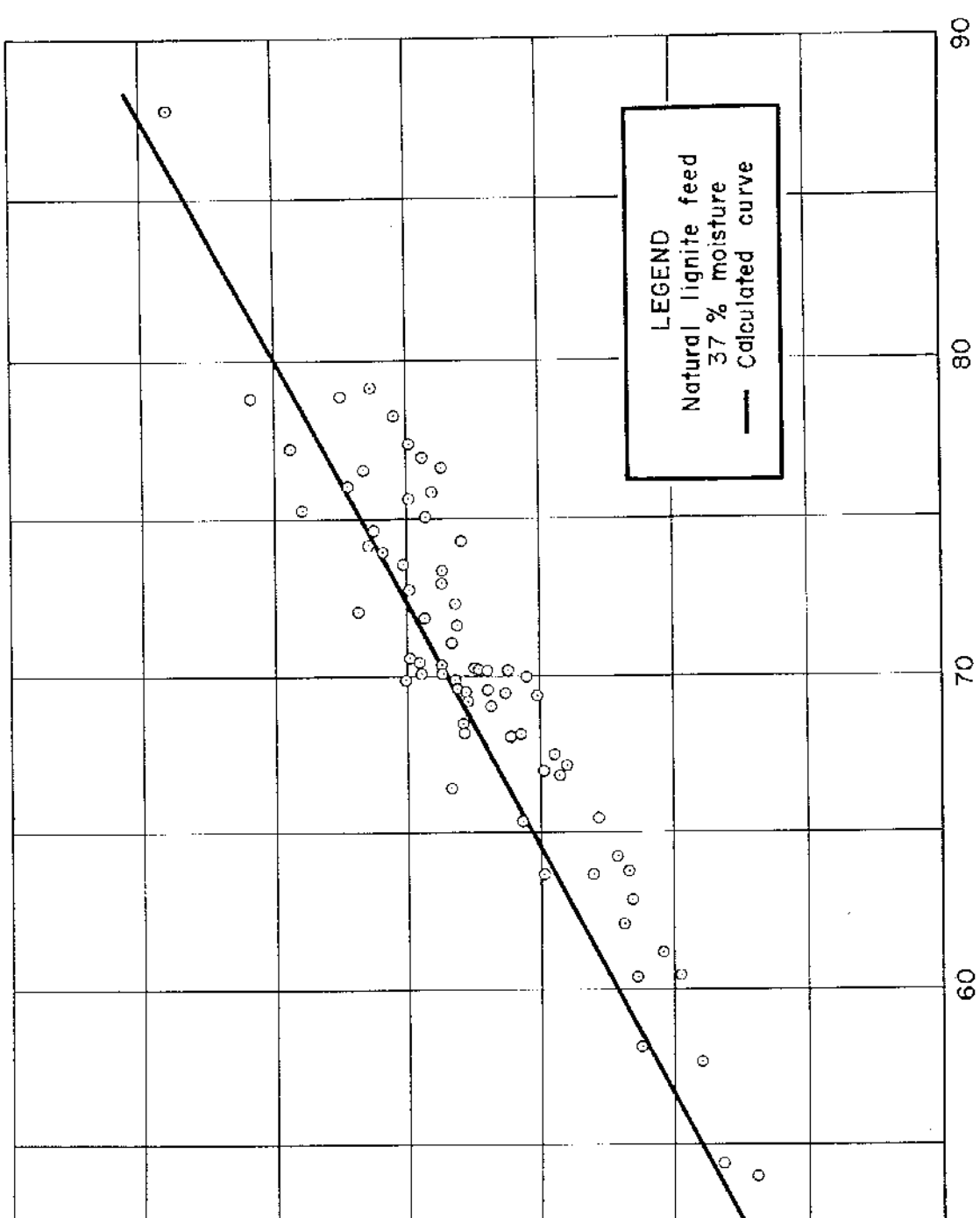

Figure 25. - Effect of percent carbon gasified on production of (H_2+CO).

Figure 26 presents a general correlation of rate of gas generation in the annular-retort gasifier as a function of heat release in the external heating furnace. Data presented in figure 26 are for the complete series of runs in the gasification pilot plant through run 17, including data from run 12 on gasification of steam-dried lignite.

This correlation plot indicates that, despite wide variation in operating conditions, the rate of gas generation was determined primarily by rate of heat release in the external heating furnace. The lower heat requirement for gasification of steam-dried lignite evidently results from elimination of part of the heat requirement for vaporization of moisture. The above correlation of rate of gas generation in terms of heat-release rate applies remarkably well over the range of operating conditions and within the limits of percent carbon gasified that were investigated. This correlation makes it obvious that the way to achieve high gasifier capacity is to increase heat-release rate in the gasifier heating furnace, if the resulting temperature levels can be tolerated at the alloy-steel heat transfer surface of the gasification retort. It is evident that the correlation could not apply to extremes of operating conditions, for example, if the lignite feed rate was raised to the point where only drying was accomplished or if an extremely high carbon gasification were sought so heat losses became excessive in relation to the heat utilized in the gasification process. At constant temperature in the heating chamber, the temperature gradient for heat transfer across the alloy tube would vary from a maximum level in the drying zone to a minimum value during gasification of the last fractions of residual carbon.

In connection with the correlation presented in figure 26, it may be pointed out why control temperatures measured in the combustion space were not satisfactory as a correlating variable for the overall correlation of gasification rate. Combustion-space temperatures are considered to give an approximate representation of temperatures at the externally heated surface of the gasification retort. In run 17, during which combustion-space temperatures were held approximately constant, rate of heat release varied from 118 to 164 million B.t.u. per hour. Steam-lignite ratio was held approximately constant during run 17, and lignite feed rate was varied only over a narrow range. Thus the changes in heat-release rate and rate of gas generation resulted in variation in the percent carbon gasified over a maximum range from 58 to 86 percent. Nevertheless all the data from this run fall in very well in the correlation plot of figure 26. In run 17, as previously discussed, the lower rates of gas generation apparently resulted from buildup of ash deposits on the inside of the alloy-steel wall of the annular retort, limiting heat transfer, and were also affected by variations in heat transfer characteristics with size distribution of the several lignites charged. When heat transfer rate became less favorable, the rate of heat release in the external heating furnace was also cut back to maintain approximately the same combustion-space temperatures.

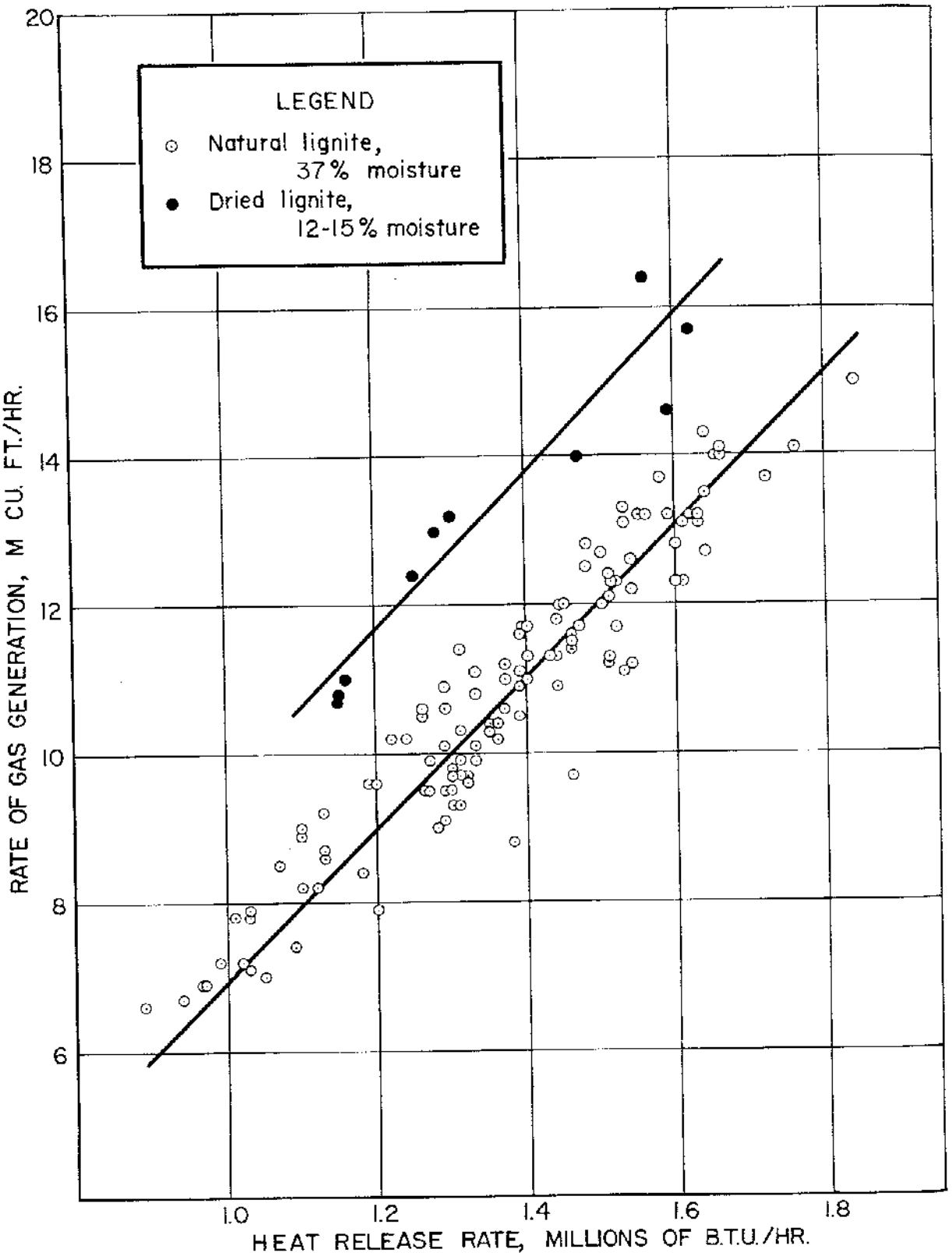

Figure 26. - Gas made versus rate of heat release in the gasifier heating furnace.