SUMMARY AND CONCLUSIONS

Experimental work is reported on the gasification of 90-percent through 200-mesh, strongly coking Sewickley-bed coal with oxygen and with steam in three temperature ranges, namely, 2,700° to 3,400° F., 1,600° to 2,000° F., and 230° to 250° F. Thirty-one runs have been made, during which 56,000 pounds of coal have been charged to the generator at feed rates of 200 to 450 pounds of coal per hour. The average results, not the optimum results, are given in the following tabulation:

Average requirements per 1,000 S.C.F. (standard cubic feet) of ($CO + H_2$) produced

Steam temperature, °F.	Coal, lbs.	Oxygen, S.C.F.	Steam, lbs.
2,904	33.3	171	81.1
1,899	37.3	326	42.5
238	42.3	394	29.1

In general, the pilot plant performed very well. Continuous operation was possible, the runs being terminated voluntarily. The percentage gasification of the coal was very satisfactory. The average percentage carbon gasified in the high-temperature steam runs was 88.9, in the medium temperature steam runs 89.9 and in the low-temperature steam runs 85.2. The oxygen requirement per 1,000 S.C.F. of (carbon monoxide plus hydrogen) produced in the high-temperature steam runs was lower than for any other continuous process known.

Refractory materials suitable for use in superheating steam to the range of 2,700° to 3,400° F. have been found to be available at a reasonable cost.

The process indicates adaptability for the production of synthesis gas on a commercial scale. The type of coal used is not critical, so that coal can be purchased on a heat-content basis. The ratio of hydrogen to carbon monoxide may be adjusted within limits. The process appears to be applicable to several industries where mixtures of carbon monoxide and hydrogen are desired.

INTRODUCTION

The Bureau of Mines, U. S. Department of the Interior, at its Morgantown, W. Va., station, operating under a cooperative agreement with West Virginia University, is conducting research and development work on the problem of producing low-cost synthesis gas directly from raw coal. This synthesis gas, consisting essentially of carbon monoxide and hydrogen, can be converted by well-known procedures into gasoline, oil, pipe-line gas, ammonia, alcohol, etc. The work described in this report covers pilot-plant-scale experiments on the gasification of pulverized coal entrained in oxygen and steam. The design of the pilot plant allows for the investigation of the relative advantages of supplying heat for the reaction of carbon with steam either by superheating the entering steam to a high temperature or by partial combustion of the coal with oxygen. Considerable general interest has been evidenced in the project, as exemplified by the many visitors to the project. Because of the interest shown in this experimental work, this progress report has been prepared to describe developments to date.

ACKNOWLEDGMENTS

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THEORETICAL CONSIDERATIONS AFFECTING DESIGN OF PILOT PLANT, INCLUDING A COMPARISON OF VARIOUS COAL-GASIFICATION SYSTEMS

According to the method of contacting the coal and the gasification agent, coal-gasification systems may be divided into three categories: (1) Fixed-bed, (2) fluidized-bed, and (3) entrainment systems. Until comparatively recently, the only systems used commercially have been fixed beds, that is, the standard water-gas set and the gas producer. Furthermore, because of the nature of coal, coke rather than coal has been the material most frequently gasified. In their search for a continuous low-cost method of coal gasification, the Germans have tried the other two systems, the fluidized-bed system being exemplified by the Winkler4/ process and the entrainment system by the Wintershall-Schmalfeldt5/ process.

4/ Morley, R. J., Winkler Generators for manufacture of Water Gas, etc.: British Intelligence Objectives Subcommittee Final Rept. 333, item 30, ca. 1946, 37 pp.

5/ Morley, R. J., The Wintershall-Schmalfeldt Process for the manufacture of Synthesis Gas at Lutzkendorf: British Intelligence Objectives Subcommittee Final Rept. 1142, item 30, ca. 1946, 33 pp.

The fixed-bed system has the decided advantage of providing a countercurrent method of gasification. As a result, a high-percentage utilization of both fuel and gasification agent is achieved, and high thermal efficiencies are obtained because the exit gas preheats the entering fuel. Furthermore, since a high relative velocity is maintained between the gasification agent and the fuel, the reaction rates are not limited as much by slow diffusion. The fixed-bed system has two chief diadvantages, namely (1) close dependence for operability on fuel and fuel-bed properties and (2) lack of temperature control in the reduction zone. Most coals have either a plastic range in which they melt and coalesce or else a range in which they disintegrate, and many have an ash-fusion temperature below the desired reaction temperature. Consequently, since these properties tend to result in poor fuel beds, it generally proves desirable to coke the coal before gasifying it and to limit the initial reaction temperature to avoid excessive clinker trouble. In turn, the heat available in the reduction zone is limited so that the temperature level drops below that necessary for reaction before the reaction is complete. Furthermore, if the usual source of fuel fails and if the operator has to buy untried fuel, the latter often is not suitable for operation at the usual throughput or produces excessive carbon losses.

The fluidized-bed system loses the advantage of the countercurrent principle with respect to the fuel unless a complicated succession of stages is substituted for the single apparatus. With respect to the gasifying agent, however, the countercurrent principle is maintained to some extent. Heat recovery from the gas stream is not so effective as in the fixed bed, for the fluidized bed operates with essentially the same temperature throughout the bed. However, this constancy of temperature makes possible good temperature control in the reduction zone. Maintenance of a satisfactory bed is difficult because of close dependence on such coal qualities as plastic range, disintegrating tendencies, and ash fusion. Agglomeration into larger particles which will drop out is a problem accentuated by coking properties or by formation of liquid slag.

The complete entrainment system has the disadvantage of being a cocurrent process, so that, as the concentration of fuel becomes low, the concentration of the gasifying agent also becomes low. As a result, a long reaction time is necessary to obtain a high percentage conversion of the reactants, and the reaction products tend to leave the reactor at a high temperature, with resultant loss of thermal efficiency. The extended area of the finely pulverized fuel available for reaction partly offsets this disadvantage. Since the solid particles are small, the relative velocity between the solid and the gas is small, so that the controlling mechanism in the reaction is diffusion of the gasifying agent through the film surrounding the particle. Since the particles are widely spaced, they may pass through the plastic range without appreciable agglomeration; furthermore, even though the ash does melt in the reaction zone, large particles are not built up. Therefore, the entrainment system tends to be independent of specific coal properties, such as plastic range, ash-fusion temperature, etc., and thus to be applicable to any coal.

This system was chosen because it would not be necessary to pay premium prices or to depend on a particular source of fuel. In either of the other two systems (fixed-bed or fluidized-bed) some preliminary treatment of the coal is customary before gasification, resulting in increased costs.

Mechanism of Coal-Particle Gasification

A physical picture of the gasification of pulverized coal completely entrained in a gasifying agent may be obtained by following the mechanism of gasification of a single coal particle. If the particle and its surrounding gas are considered to be spheres, the ratio of the diameter of the gas sphere to that of the particle is about 37:1 at atmospheric pressure. Hence, the gas space surrounding the particle is large, and good mixing is an important factor in promoting the reaction. If the coal contains 70 percent carbon and 10 percent ash, if all the volatile matter and 90 percent of the carbon are gasified, and if the volume change is proportional to the weight change, then the ratio of the diameter of the particle after the reaction to that before the reaction would be expected to be 0.55.

For a 200-mesh particle falling at its terminal velocity in a gas (CO, CO_2, H_2O) at 2,000° F., the Reynolds number is approximately 0.2, so that Stokes law applies. The terminal velocity or the maximum relative velocity between the gas and the particle falling under the influence of gravity is 0.14 ft./sec.[6] Meyers[7] has shown that the major resistance to the reaction between carbon and carbon dioxide is diffusional below relative velocities of 0.7 ft./sec. and chemical above that velocity when the reaction temperature is 1,800° F. The major resistance to reaction in entrainment coal gasification may be concluded to be diffusion; and, therefore, an increase in the temperature level will not produce a large increase in reaction rate. Since the relative velocity between the coal particle and the gas is low, turbulence is of doubtful value in promoting the reaction, for the coal particles will move with the gas. One effective method of obtaining a high relative velocity between the coal particle and the gas would be to cause the particle to move in a spiral path so that a centrifugal force several times that of gravity is obtained; that is, a much greater relative velocity is obtained than that caused by turbulence only.

When a 200-mesh coal particle enters a region at 2,000° F., it is heated rapidly through the plastic range, the volatile matter being distilled off and forming an envelope around the particle. Hence, the initial reaction with oxygen is combustion of the volatile matter. Most bituminous coals contain enough volatile matter to react with most of the oxygen; and, since gaseous combustion reactions are rapid, most of the oxygen disappears in a short length of the reactor. Consequently, the reactions occupying most of the reactor space are those of steam and carbon dioxide with the fixed carbon of the coal. Very high initial temperatures are not observed, because the steam present limits this temperature as in the usual operation of a fixed bed gas producer.

6/ Dalla Valle, J. M., Micromeritics: Pitman Publishing Corp., New York. 2d ed., 1948, p. 20.

7/ Meyers, L., Trans. Faraday Soc., vol. 34, 1938, p. 1056.

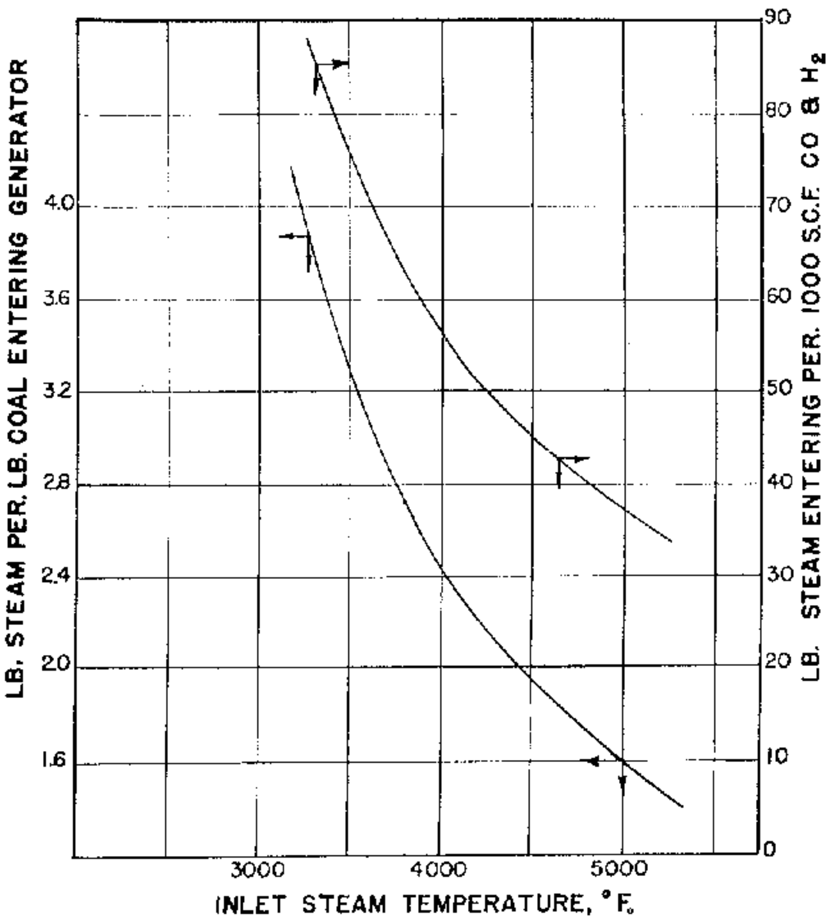

Figure 1. - Calculated steam requirement for gasification of standard Sewickley coal, using no oxygen.

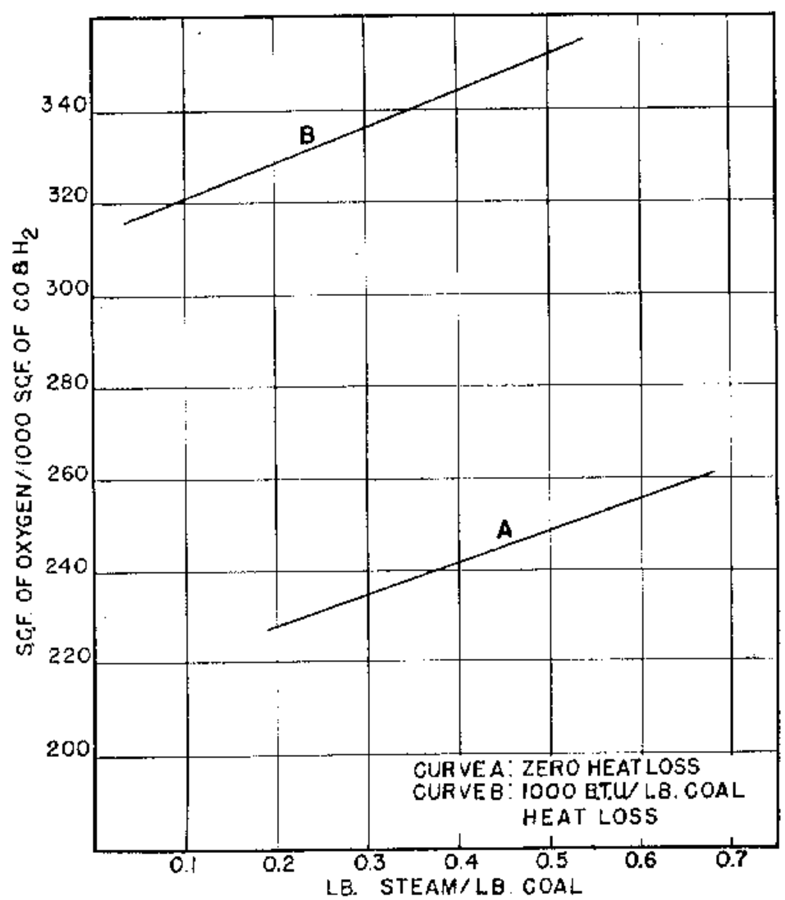

Figure 2. - Calculated effect of steam:coal ratio on oxygen requirement for gasification of standard Sewickley coal.

Heat Supply

In gasification work in general, the reactions of carbon dioxide and steam with carbon[8] require that heat be supplied to the process. Heat may be supplied in three ways, namely (1) by being transferred through a retaining wall, (2) by being generated within the process, and (3) by entering as sensible heat in the entering reactants.

If retaining walls are used and the reactants are externally heated two difficulties are encountered: (1) Using refractory materials, capable of withstanding high temperatures, the heat-transfer rate is low because of the resistance offered by the wall and two stagnant gas films; (2) if high heat-transfer rates are sought by the use of metal walls, the reaction is slowed, since the temperature of the metal must be kept comparatively low - under 2,000° F.

Heat may be supplied for the process by preheating the entering steam to a high temperature. Figure 1 shows the calculated steam requirement (steam:coal ratio and steam: $(CO + H_2)$[9] ratio) for various entering steam temperatures when all the heat for the strongly endothermic reaction of carbon with steam or CO_2 is supplied as sensible heat of the entering steam. The assumptions used in making these calculations are: (1) 90 percent of the carbon in the coal is gasified, (2) the products reach equilibrium with respect to the water-gas shift reaction[10] at an exit temperature of 2,000°F., and (3) there is no radiation heat loss from the apparatus. The gas was assumed to contain CO, H_2, CO_2, and H_2O plus 1 pound of methane and 2 pounds of ethylene per 100 pounds of coal. The analysis of the strongly coking Sewickley bed coal used as standard is as follows on the as-received basis:

	Percent		Percent
Moisture.......	0.7	Oxygen.......	7.3
V. M.	35.7	Sulfur.......	2.3
F. C.	50.6	Nitrogen.....	1.4
Ash............	13.0		Btu/lb.
Carbon.........	71.1	Gross heating value	12,770
Hydrogen.......	4.2	Net heating value	12,360

It is seen from figure 1 that, to supply all the heat for the reaction in the entering steam, either an excessively high steam temperature is required, or steam in an excessively large amount is required.

Heat may be supplied for the process by the partial combustion with oxygen of coal or the volatile matter from coal. Figure 2 shows the oxygen

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8/ At 2,000°F.,

$C + CO_2 = 2\ CO$ - 5,860 B.T.u./lb. of C reacting.

$C + H_2O = CO + H_2$ - 4,100 B.t.u./lb. of C reacting.

9/ Gas volumes are given in standard cubic feet (S.C.F.) measured at 60°F., 30 in. Hg, dry.

10/ At 2,000°F., $CO + H_2O = CO_2 + H_2$ + 21, 500 B.t.u./lb. mol.

requirement (oxygen:($CO + H_2$) ratio) for various steam:coal ratios when 240°F. steam is used and when the same assumptions are used as in the previous calculations. In this case, the oxygen requirement of the process is high, and the oxygen is the most expensive ingredient per unit of $CO + H_2$ produced. The best method of supplying heat for the process is the one that costs the least. An economic balance between heat supply as sensible heat of the entering steam and heat supply as heat from combustion with oxygen will determine the best operating conditions. Figure 3 shows the oxygen requirement for three steam:coal ratios for various steam temperatures.

Extent of Reaction

Within the experimental accuracy of the German data, 11/12/13/ the water-gas shift reaction reached equilibrium at the exit-gas temperature. The reactions of carbon dioxide and steam with carbon, however, were far from equilibrium in all coal-gasification systems. In general, if a low steam:coal ratio is used in a gasifier, the gasifying agent is almost exhausted at the exit end of the apparatus. If a high steam:coal ratio is used, however, the exit gas still contains a fairly high concentration of gasifying agent. It could be expected from the mass-action effect that a greater carbon gasification and a lower exit-gas temperature would obtain for high steam:coal ratios in entrained coal-gasification systems.

The exit-gas temperature is strongly influenced by the reaction rate at the exit end of the reactor. In a cocurrent process for the gasification of pulverized coal completely entrained the exit end of the reactor contains the lowest concentrations of fuel and gasifying agent, as well as the lowest temperature - all conditions which decrease the reaction rate. It is to be expected, then, that equilibrium will not be attained and that considerable energy which could be converted to chemical energy according to the equilibrium relations will actually be left as sensible heat in the exit gas. Figure 4 shows the calculated oxygen requirement for various exit-gas temperatures when the Sewickley coal chosen as standard is used. It is seen that the lower the exit-gas temperature, the more efficient is the process. The exit-gas temperature can be influenced by the shape of the apparatus. If the reacting mass flows progressively through a tube, a temperature gradient is established along the tube as the reaction takes place, resulting in a lowered exit-gas temperature and, hence, a greater extent of reaction. If the reacting mass is mixed by turbulence in the reactor, however, short-circuiting raises the exit-gas temperature and decreases the extent of the reaction.

11/ See footnote 4.

12/ Atwell, H. V., Koppers Powdered-Coal-Gasification Process: Joint Intelligence Objectives Agency, FIAT Report 1303, Washington, D. C., September 1947, 51 pp.

13/ Atwell, H. V., Gumz Powdered-Coal-Gasification Process: Joint Intelligence Objectives Agency, FIAT Report 1304, Washington, D. C., September 1947, 53 pp.

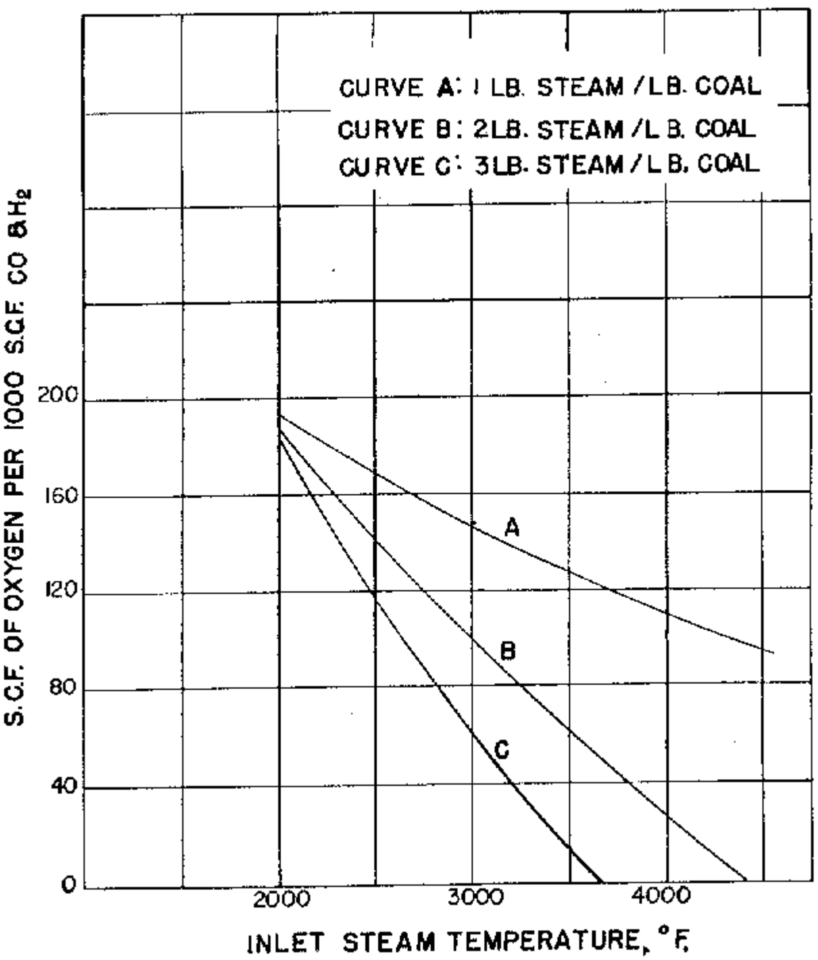

Figure 3. - Calculated effect of entering steam temperature on oxygen requirement for gasification of standard Sewickley coal.

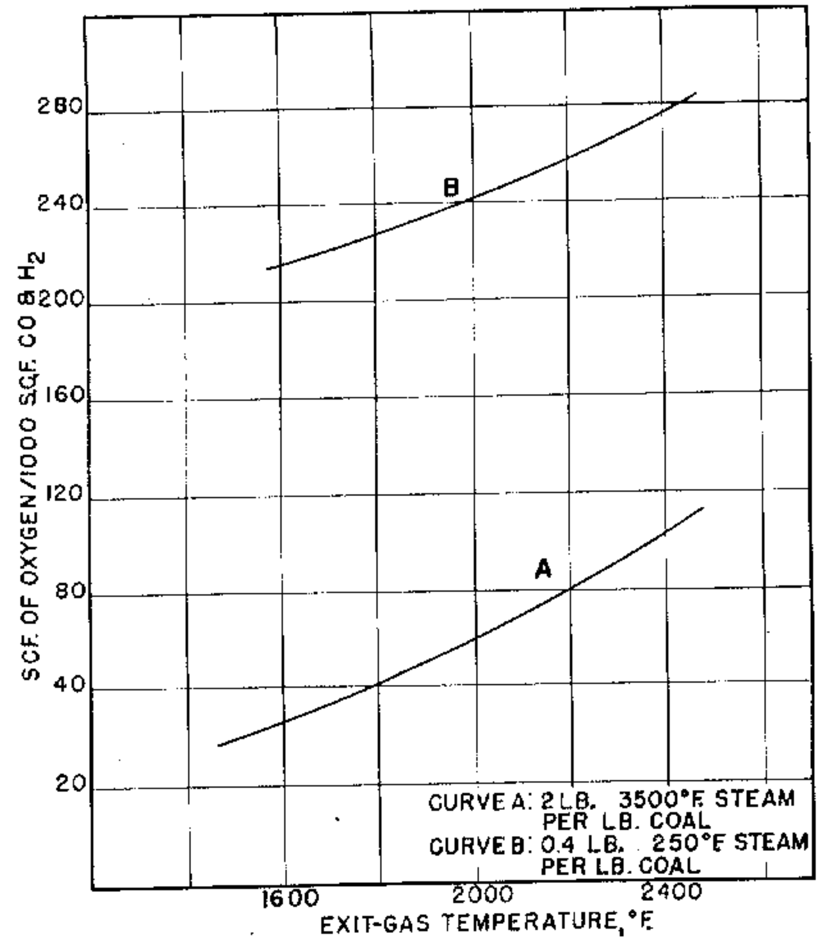

Figure 4. - Calculated effect of exit-gas temperature, °F., on oxygen requirement.

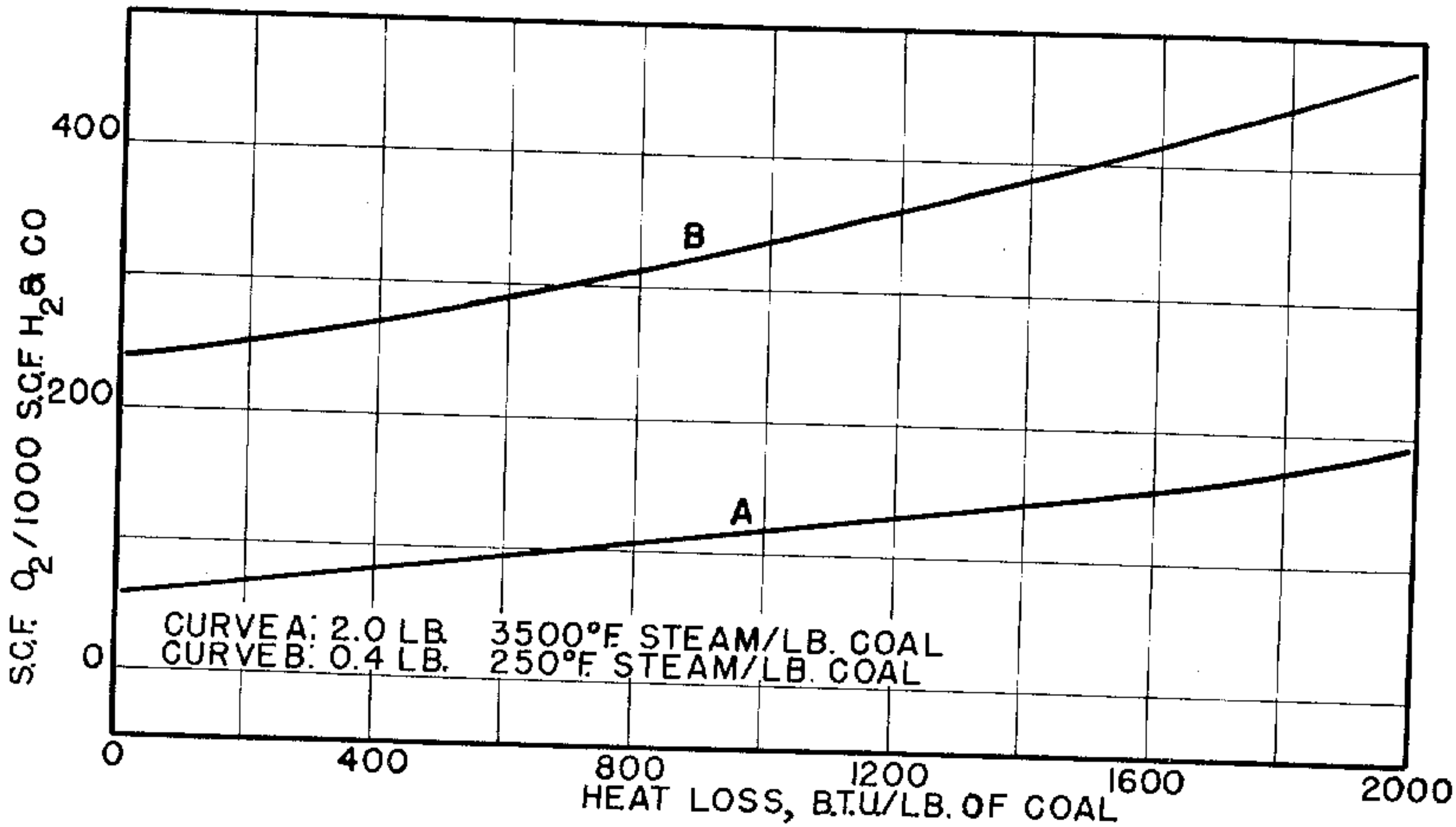

Figure 5. - Calculated effect of heat loss on oxygen requirement.

It is seen in figure 5 that the oxygen requirement per unit of $CO + H_2$ produced increases with increase in heat loss by radiation from the gasifier to the surroundings. Since heat loss by radiation is proportional to surface area, it will increase with the ratio of area to volume or length to diameter of the gasifier. Consequently, a balance or compromise is necessary between the optimum shape and size of reactor for orderly progression of the reacting materials and that for low heat loss.

Reactor Design

From the foregoing discussion, the following principles obtain for the design of a reactor for the gasification of pulverized coal by the entrainment system. The coal particles should move in a spiral path so that the relative velocity between the coal particles and the gas is maintained at as high a value as possible, the object being to reduce the resistance of the gas film and increase the transfer rate of the gasifying agent from the gas stream to the carbon particle. A large reaction space or a long contact time should be provided to allow the reactions of CO_2 and steam with carbon to proceed when the concentrations are low and the rate is, hence, reduced. All the reactants should enter at one end of the reactor and flow uniformly through the reaction space to reduce the possibility of short circuiting and to obtain the maximum conversion. The ratio of surface to volume of the reactor should be as low as is consistent with the previous requirements, so that the heat loss will be held to a minimum since any heat loss produces a corresponding decrease in the quantity of the desired products. These principles influenced the design of the reactor to be described in this report.

Since a study of the use of high-temperature steam as a source of part of the heat supply for the process was also desired, the necessary equipment for producing extremely hot steam was designed.

Original Design and Flow Sheets

The theoretical considerations that guided the experimental approach and influenced the generator design for the pilot-plant work at Morgantown have been set forth in the previous section. A description of the pilot plant and the flow sheet for the process have been published previously.[14] The construction of the first experimental unit, which will be described more completely later in this report, was substantially as published; modifications made were largely to secure simplification in construction or to conserve space.

The process shosen - gasification of finely pulverized coal entrained in oxygen and superheated steam - appeared to be one that would not require carefully selected or premium fuels. The pilot-plant design, and particularly

14/ Schmidt, L. D., McGee, J. P., and Slone, M. C., A Pilot Plant for Gasifying Powdered Coal entrained in Oxygen and Steam: Chem. Eng. Prog. vol. 44, No. 10, October 1948, pp. 737-744.