I. DEVELOPMENT OF PROCESS IN LABORATORY UNIT OPERATING ON ENTRAINED POWDERED COAL

INTRODUCTION

The process development described in this report was carried out at the Morgantown Station of the U. S. Bureau of Mines in cooperation with West Virginia University. An experimental laboratory-scale pilot unit to gasify coal dust in entrainment was constructed on the Campus of the University in Morgantown.

The enormously accelerated demand for liquid and gaseous fuels and utilization of our immense coal resources require that a low-cost process be developed for the gasification of a wide variety of coals. Such a process would be a major factor in lowering the cost of synthetic liquid fuels. None of the processes developed in Germany prior to and during World War II, such as the Winkler process[5][6][7], the H. Koppers dust gasification process[8], the Wintershall-Schmalfeldt process[9], etc., meet all the requirements for a low-cost process under American conditions. Nor has any other process designed and experimentally operated at atmospheric pressure during the post-war period in this country appeared to fulfill all of the necessary conditions for a low-cost process.

The process described in this paper, based on the continuous production of synthesis gas by "downdraft" entrainment of powdered coal in oxygen and superheated steam, has been in operation for more than 3 years,[10] being, as far as is known, the first pulverized-fuel gasification unit in this country capable of continuous operation. Operated at feed rates up to 45 pounds of coal per hour, corresponding to an hourly throughput rate of about 33 pounds of coal per cubic foot of generator volume, the process is well suited for testing various types of fuels for their utility in

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[5] Gas-Making Processes: American Gas Association Report, Oct. 1945, 160 pp.

[6] Winkler Gasification Process: Coke, vol. 8, 1946, pp. 155-8.

[7] Newman, L. L., Oxygen in the Production of Hydrogen or Synthesis Gas: Ind. Eng. Chem., vol. 40, 1948, pp. 559-82.

[8] Atwell, H. V., Koppers Powdered-Coal Gasification Process: FIAT Final Report No. 1303, P.B. 85165, Joint Intelligence Objectives Agency, Washington, D. C., Sept. 1947.

[9] Morley, R. J., The Wintershall-Schmalfeldt Process for the Manufacture of Synthesis Gas at Lutzkendorf: BIOS 1142, 1945.

[10] The first run was made on May 26, 1947 (see Run No. 1 in the Appendix).

making synthesis gas on a commercial scale. Owing to its simplicity and ease of control, the apparatus can be used for much-needed fundamental research on the kinetics and mechanism of powdered-fuel gasification.

A synthesis gas containing 90 percent $CO + H_2$ and about 7 percent CO_2 can be made from low-grade coking or noncoking coals, and the residue obtained can be recycled for further gasification. Gas-output rates as high as 600 cu. ft. $CO + H_2$ per cubic foot generator volume per hour have been obtained, with indications that even this unusually high rate would be exceeded considerably when coals of younger geologic age are gasified under expediently chosen operating conditions.

A study was begun of the effect of operating variables on which the efficiency of gasification and the cost of synthesis gas depend to a large extent. The most essential of these variables are the type of coal and its fineness of pulverization, coal feed rate, oxygen-to-coal and steam-to-coal ratios, and steam temperature. Optimum process efficiency should result from the most favorable combination of these operating conditions. As it was found advantageous to operate with excess carbon, which at high coal throughput rates results in appreciable quantities of an extremely fine and fluffy carbonaceous residue, it is essential to make a study of this byproduct of gasification from the standpoints of the economy of recycling and utilization as generator fuel or other commercial uses.

THEORETICAL

In table 1 are listed the five fundamental reactions concerned in the gasification of coal. Based upon data[11] published by the National Bureau of Standards, the reaction heats evolved or absorbed have been calculated for three different temperatures, corresponding to 1,300°, 1,400°, and 1,500° Kelvin, both reactants and reaction products assumed to be at these respective temperatures. This expresses the amount of heat involved much nearer the temperatures at which the reactions take place than the figures commonly used in various textbooks, which are based on both reactants and products being at 60° F. In the table, water is assumed to be in the gaseous state, and the carbon is assumed to be in the state of graphite. Exothermic of endothermic heats are shown by a plus or minus sign, respectively.

TABLE 1. - Heats evolved or absorbed in gasification reactions

Reactions	Reaction heat, in B.t.u. per lb. mol 1,880° F. (1,300° K.)	2,060° F. (1,400° K.)	2,240° F. (1,500° K.)
1. $C + 1/2O_2 = CO$	+ 48,982	+ 49,277	+ 49,581
2. $C + O_2 = CO_2$	+170,021	+170,109	+170,199
3. $C + CO_2 = 2CO$	- 72,056	- 71,557	- 71,035
4. $CO + H_2O = CO_2 + H_2$	+ 13,716	+ 13,331	+ 12,958
5. $C + H_2O = CO + H_2$	- 58,340	- 58,226	- 58,077

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[11] Wagman, D. D., Kilpatrick, J. E., Taylor, W. J., Pitzer, K. S., and Rossini, F. D., Heats, Free Energies, and Equilibrium Constants: National Bureau of Standards Res. Paper RP1634, 1945.

In gasifying coal entrained in steam and oxygen at these temperatures, the oxygen reacts with carbon in the coal to form carbon monoxide and carbon dioxide, the ratio of the two depending essentially on gasification conditions, including the temperature of the generator chamber, the ratio of oxygen injected to coal charged, and the ratio of steam to coal. The exothermic combustion reactions furnish the heat required for the endothermic reaction of the steam with the remaining carbon, to form hydrogen and carbon monoxide, the essential constituents of synthesis gas.

Let it be assumed, for the sake of simplicity, that the oxygen used is just sufficient to burn part of the carbon with which it reacts to CO (without forming any CO_2), and the heat developed by Reaction 1 is absorbed entirely by Reaction 5, consuming the stoichiometric amounts of steam and carbon, namely 0.854 mols of each, so that the balance of heat is nil.[12] Then, provided that there are no heat losses, and Reactions 1 and 5 are assumed to proceed to completion at 2,240° F., each pound of carbon in the coal would require 8.5 cu. ft. (0.72 lb.) of oxygen[13] and 0.69 lb. of steam for the reactions. The upper limit of yields would thus be 31.5 cu. ft. of CO plus 14.5 cu. ft. of H_2 per lb. of carbon, or a total of 46 cu. ft. of gas containing 31.5 percent H_2 plus 68.5 percent CO, the CO:H_2 ratio being 2.2. Thus, with an oxygen-to-carbon input ratio of 0.72 lb. O_2 per lb. of C and a steam-to-carbon input ratio of 0.69 lb. H_2O per lb. of C, the theoretical oxygen requirement is 185 cu. ft. per 1,000 cu. ft. of CO + H_2. Actual gasification yields are less, of course, because of heat losses and incomplete reactions.

In the early tests with low coal throughputs, the heat losses were so excessive in the internally heated gas generator of the size used (6 inches I.D. and 82 inches length) that about twice the above oxygen input was required per 1,000 cu. ft. of synthesis gas produced in order to counteract the heat losses and maintain the generator temperature above 2,000° F. Furthermore, it was necessary to use a very low steam input, about one-eighth to one-tenth of the steam-to-carbon input ratio calculated above, otherwise the generator temperature would have dropped, and the CO_2 content of the make-gas would have gone up to 20 to 30 percent. By lowering the steam input and increasing the coal throughput, it was possible later to make a low CO_2 (6 to 7 percent) synthesis gas without resort to an excessively high oxygen input.

For a high synthesis gas yield per unit weight of coal charged, it is essential that the heat losses from the generator due to radiation, convection, and conduction be held to a minimum. Expressed as heat units lost per unit volume of generator chamber or per unit volume of synthesis gas produced, small-scale generators compare unfavorably with commercial-scale generators. The heat losses from the latter are lower, as their surface area exposed per unit volume of generator chamber is considerably less. As a result, large-scale generators require much less insulation. Conversely, there is a limit beyond which it is not feasible to increase the insulation around small generators.

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[12] Note that 85.4 percent of 58,077 B.t.u. (reaction 5) equals 49,581 B.t.u. (reaction 1).

[13] All gas data refer to S.T.P. or 60° F., 30" Hg, dry.

For the same reason, if small generators are run much below their maximum capacity, the B.t.u. loss per MCF of gas produced may be augmented considerably. Thus, by increasing the coal throughput near the maximum capacity of the generator, it was possible to reduce the heat losses to a minimum and thus improve the yield and quality of synthesis gas.

However, elimination of heat losses alone is not sufficient for the effective gasification of coal, the objective being to increase the percent gasification of carbon in the coal charged, although a complete elimination of a carbonaceous residue seems to be impractical and uneconomical. Based upon figures published by P. O. Rosin[14] in his aerodynamic studies of the combustion of coal, theoretically each volume of coal must react with about 2,700 volumes of oxygen to form CO_2, or with 1,350 volumes of oxygen to form CO, or with 1,350 volumes of steam to form $CO + H_2$. Thus, large volumes of gasifying media must be conducted to the coal surface, and large volumes of gaseous products must be transported away from it. When steam reacts with carbon, the volume of product gases is twice the volume of the steam. In order to facilitate mass transfer rates through the gaseous films surrounding each coal particle, considerable mixing and turbulence is required in the generator chamber.

It would be desirable to have a considerable difference between the relative velocities of coal and gases. Rosin suggests the injection of the gasifying oxygen and steam at a high velocity and the use of a pulverized-coal burner provided with a number of orifices, so that numerous small eddy currents would form as the coal particles leave the nozzle. These side eddies plus secondary gasifying medium divided into many separate streams and blown in with high velocity at right angles to the direction of the flame should provide a high degree of turbulence. In the experimental work to be described, considerable attention was paid to these aerodynamic principles in order to obtain the necessary turbulence and relative velocity differences, although the validity of the aerodynamic theory has not yet been confirmed.

According to the investigations of W. Gumz,[15] it is desirable that an excess of carbon be present with a deficiency of gasifying agents. This should result in more $CO + H_2$ and less CO_2 in the gas.

Of considerable importance from the standpoint of gasification efficiency is the velocity at which the reactants and the synthesis gas formed flow through the generator tube toward the outlet carrying fly ash and ungasified carbonaceous residue along in entrainment. For any given tube diameter and length there is a maximum velocity (or minimum residence time) for the reactants and products that should not be exceeded. At higher velocities, and correspondingly less contact time, the percent of carbon gasified decreases, with consequent increase in production cost. Thus, it is to be

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[14] Rosin, P. O., Fuel in Science and Practice: Vol. 15, 1936, pp. 136-48.

[15] Gumz, W., Handbuch der Brennstoff und Feuerungstechnik, Springer-Verlag, Berlin, 1942. Revised edition in English now in preparation.

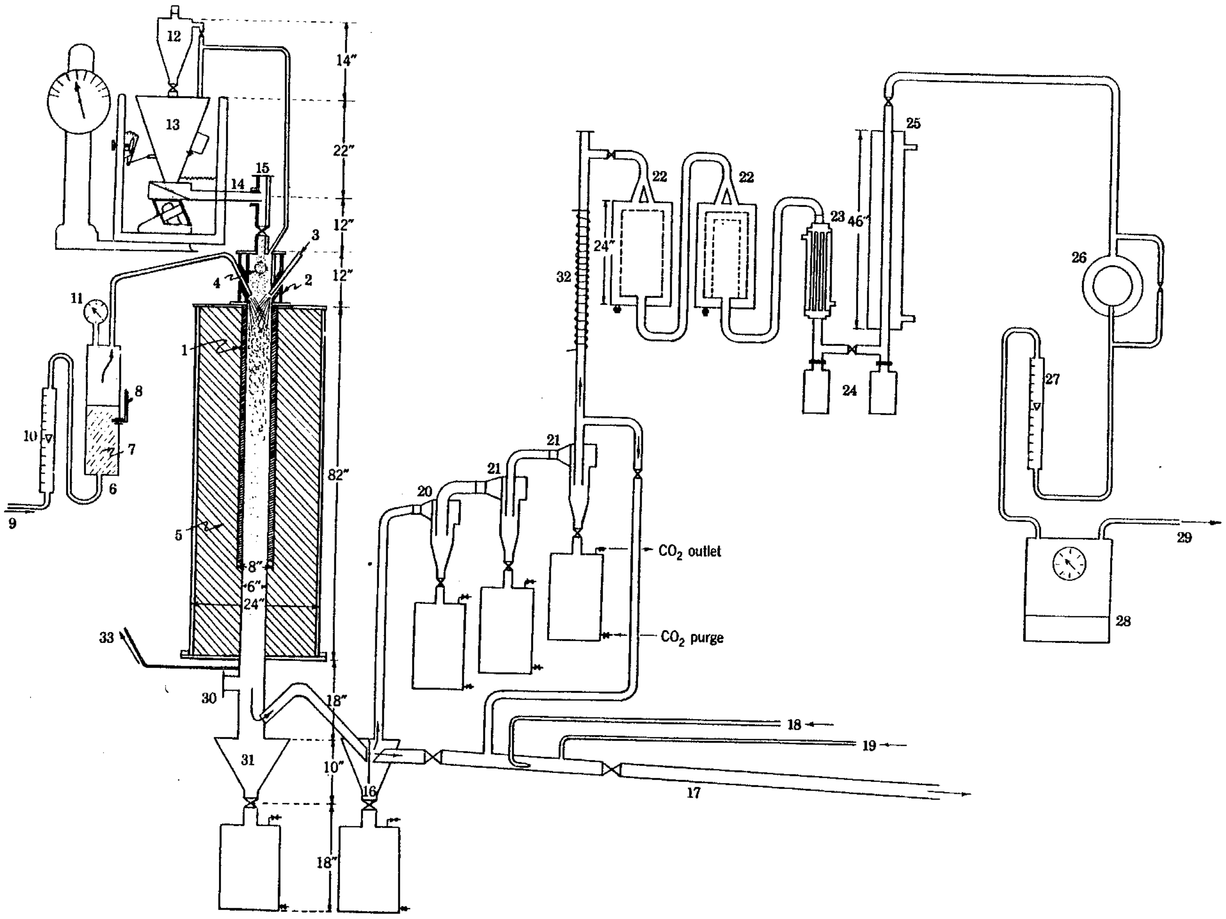

Figure 1. - Flow diagram of laboratory unit in second stage of development.

1. Silicon carbide generator tube.
2. Water-cooled jacket.
3. Oxygen jets.
4. Burner ports.
5. High-temperature insulating brick.
6. Steam-to-oxygen ratio controller.
7. Water.
8. Thermometer.
9. Oxygen.
10. Flow indicator.
11. Pressure gage.
12. Charge hopper.
13. Syntron hopper and vibratory feeder.
14. Feed tube.
15. Rupture disk.
16. Knockout chamber.
17. 2-inch ejector pipe.
18. Steam to jet.
19. Water.
20. 3-inch Aerotec tube.
21. 2-inch Aerotec tube.
22. Fiber-glass filters.
23. Tubular condenser.
24. Condenser water.
25. Secondary condenser.
26. Exhauster.
27. Rotameter.
28. Dry meter.
29. To gas holder.
30. Rupture disk.
31. Slag and residue collector.
32. Heating coil.
33. Gas sample to instruments.

expected that the tube diameter and length determine the maximum economical throughput of coal.

APPARATUS AND OPERATING PROCEDURE

The process development, which extended over a period of about two years, may be divided into three stages.

Gasification in a 3-inch tube with wet purification system

The first gasification unit built consisted of a 4-foot long silicon carbide tube of 3-inch I. D. placed concentrically inside a 6-inch I. D. silicon carbide tube. Powdered coal was charged at a rate of 1 to 5 pounds per hour from a hopper, by means of a screw conveyor and an electrically vibrated throat, into the middle of two impinging oxygen-steam jets located in a water-cooled head in contact with the top of the inner tube. The fine coal particles entrained in the turbulent stream were partly gasified at about 2,000° F. and were carried downward by the gases to the bottom of the 3-inch I. D. chamber. The small amount of slag formed, together with most of the ash and ungasified carbon, fell into the sump at the bottom of the standpipe below the generator. The gaseous products carrying small particles of residue were exhausted upward through the 1-inch wide annular space between the inner and outer tubes. The gases passed from the top of the annulus to the cooling, scrubbing, and purification equipment before being metered and stored in a gas holder.

It was expected that the annular space surrounding the generator chamber would act as high-temperature insulation. However, the soundness of this principle could not be proved owing to the excessive heat losses (B.t.u. per MCF of gas produced) due to the small capacity of the 3-inch I. D. inner chamber, which did not allow coal throughputs higher than 5 pounds per hour. Owing to the high heat losses through the 8-inch thick insulation around the outer silicon carbide tube, the temperature in the reaction chamber could not be maintained above 1,800° to 2,000° F. As a result, the gas yield per unit weight of coal was low, and the carbon dioxide content of the make gas ranged from 15 to 30 percent.

Gasification in a 6-inch tube with dry residue recovery system

To increase the throughput of the generator and lessen thereby the heat losses per unit volume of gas produced, it was decided to enlarge the gasification chamber by removing the 3-inch I. D. inner silicon carbide tube, which resulted in a plain 6-inch I. D. tube 82 inches long without any surrounding annular space. The generator thus modified and the completely altered dust-purification train are shown in the flow diagram in figure 1, representing the second stage of development. The most important advantage gained was a much larger gasification chamber, which permitted coal throughputs of 30 to 50 pounds per hour. This resulted in lower heat losses per cubic foot of gas made. Also, former troubles of frequent clogging of the annular space by residual dust have been eliminated.

An essential change was from the wet to the dry purification system, comprising a "dust collector" below the generator, a "knockout chamber", three "Aerotec" tubes, two "fiber-glass filters" of special design, followed by a multiple-tube and a single tube condenser, a Roots-Connersville type exhauster, a rotaflow meter, and a dry gas meter. Particles of slag and coarse residue dropped by gravity into the "dust collector," residue of medium coarseness was removed by impact against a baffle plate in the "knockout chamber," whereas the fine carbonaceous residue and fly ash were removed in the "Aerotec" tubes and "fiber-glass filters" in series. The residue collected in each unit was released through a blast-gate valve into a 2-cubic foot capacity drum, which could easily be replaced when filled to capacity by an empty drum.

In case of the so-called "short-cut" runs when measurements of the gasification residue and output of the make gas were not required, the generator could be run as usual and the products "ejected" through a 2-inch pipe from the knockout chamber to the outside. During the ejection, steam and a stream of water helped to keep the ejector pipe cool and prevented its clogging by residue. In "complete" runs, the ejector pipe was closed, and the products were passed through the entire train.

The dry purification system entailed several advantages over wet purification, such as complete recovery of carbonaceous residue and fly ash, direct measurement of undecomposed steam recovered in condensers, and retainment of the CO_2, much of which was formerly washed out of the gas. This permitted analysis of the process by material and heat balances.

Another important change was the substitution of a "Syntron" vibratory feeder for the former screw feeder, which made the feed rate somewhat more uniform. The pulverized coal was dropped into the water-cooled generator head from the end of a 2-inch diameter tubular trough vibrated by a pulsating DC current. Several difficulties, owing to the tendency of very finely powdered coal to pack at the bottom of the feed hopper, were eliminated by agitation of the coal in the hopper with a small amount of inert gas (nitrogen) introduced at its throat. The nitrogen thus used was then passed into the generator head and served the additional purpose of preventing the upward surge of steam and oxygen toward the mouth of the feed trough.

A scale on which the feeding mechanism rested allowed a check on the feed rate by weighing the amount charged in each 3-minute period. Thus, it was found that fluidization of the charge in the feed-hopper improved the feed rate, so that its variations over 3-minute intervals dropped from ±25 to ±5-10 percent. Yet, even this variation proved to be detrimental, in spite of constant feed rates over 1-hour or 30-minute periods. Even slight variations in the feed rate caused undue fluctuations in the generator back pressure, which, in turn, induced further irregularities in the feeding. Thus, the sensitivity of this feeder to variations in back pressure created a vicious cycle. For this reason, the Morgantown Station of the Bureau of Mines developed a new type of pneumatic feeder, which proved to be superior to the vibratory feeder. The latter was replaced, therefore, after a few months' use, by the pneumatic feeding system.

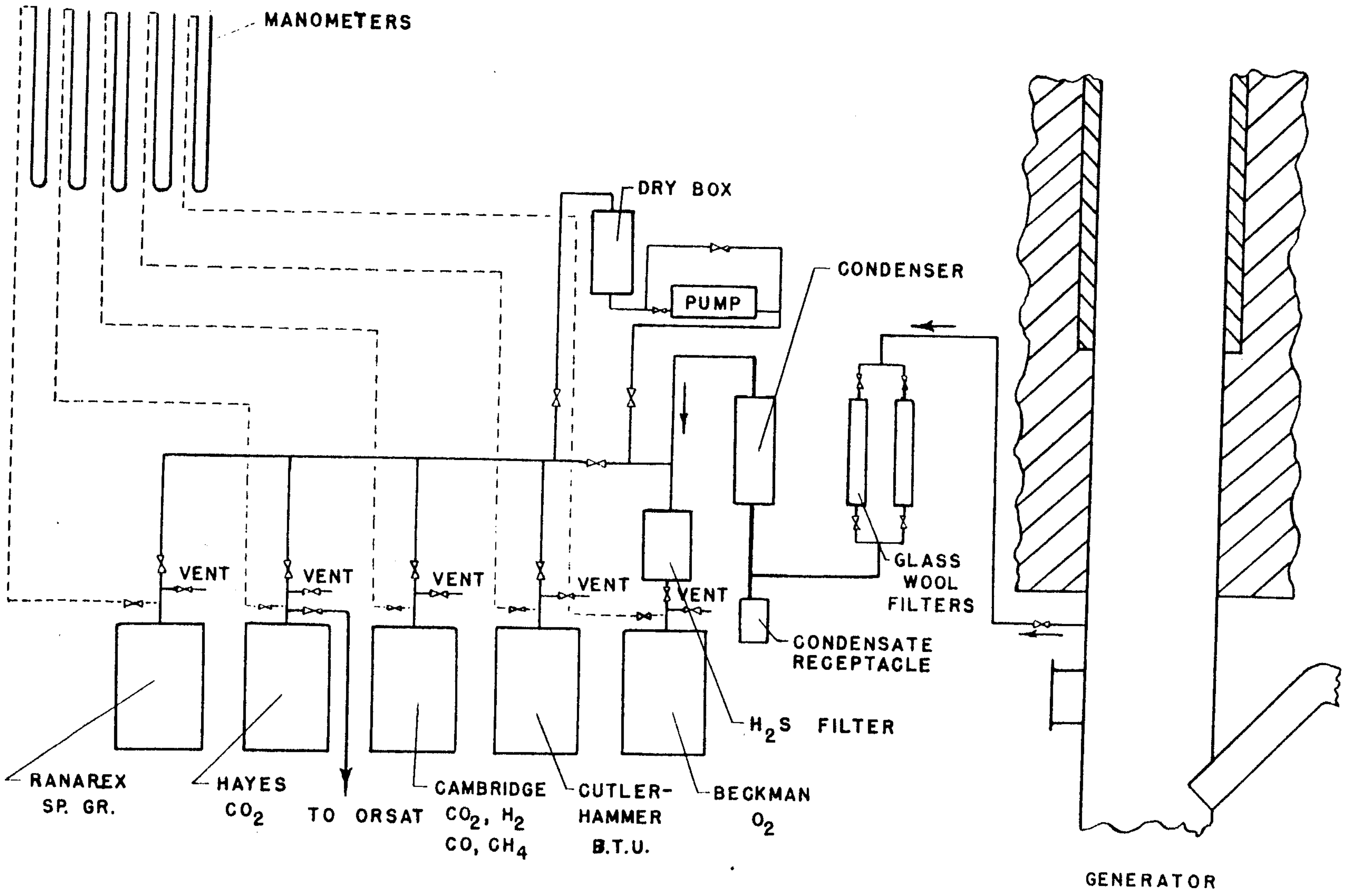

Figure 2. - Gas-sampling and testing arrangement in laboratory unit.

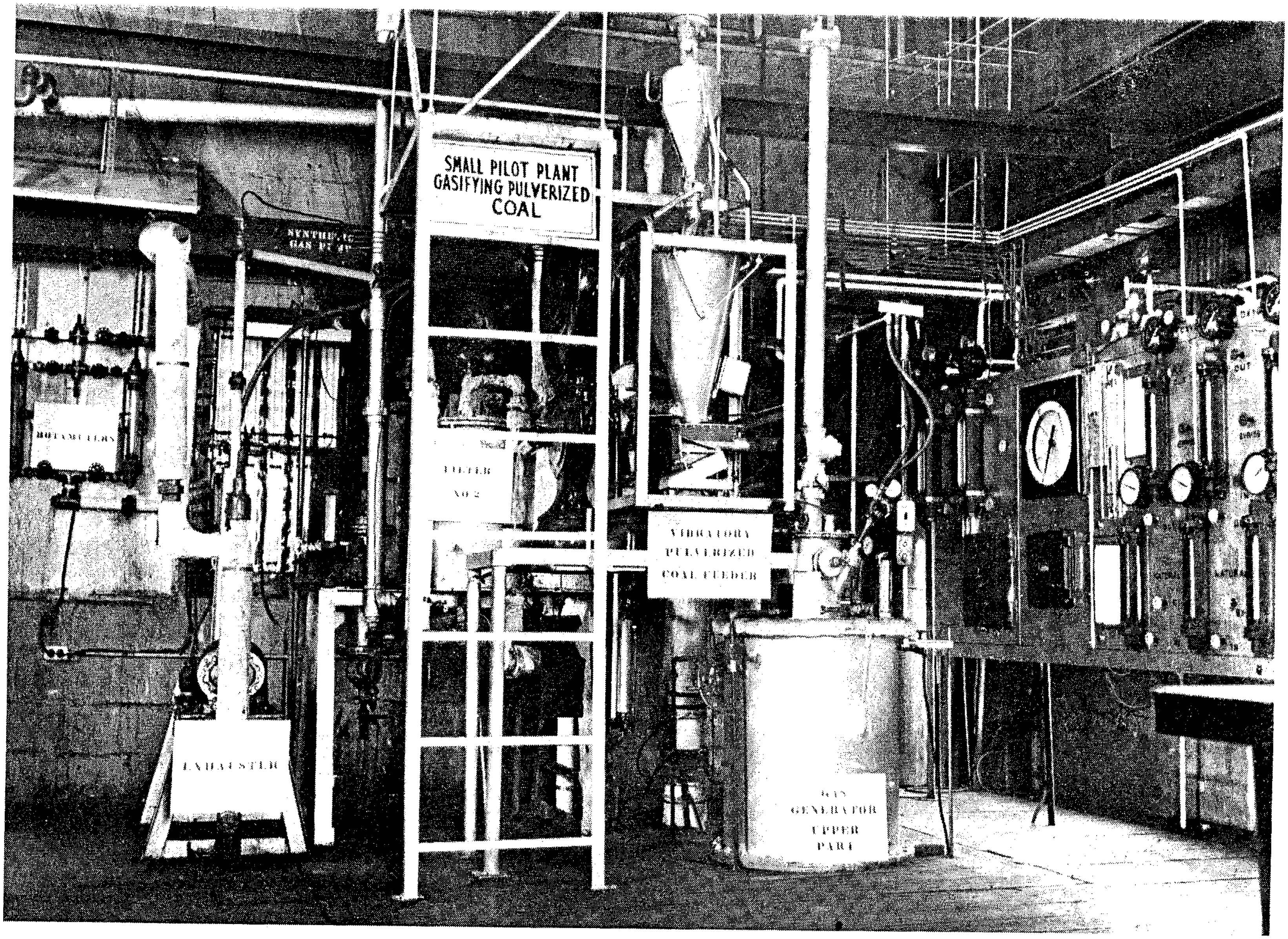

Figure 3. - Laboratory unit (upper floor) in second stage of development.

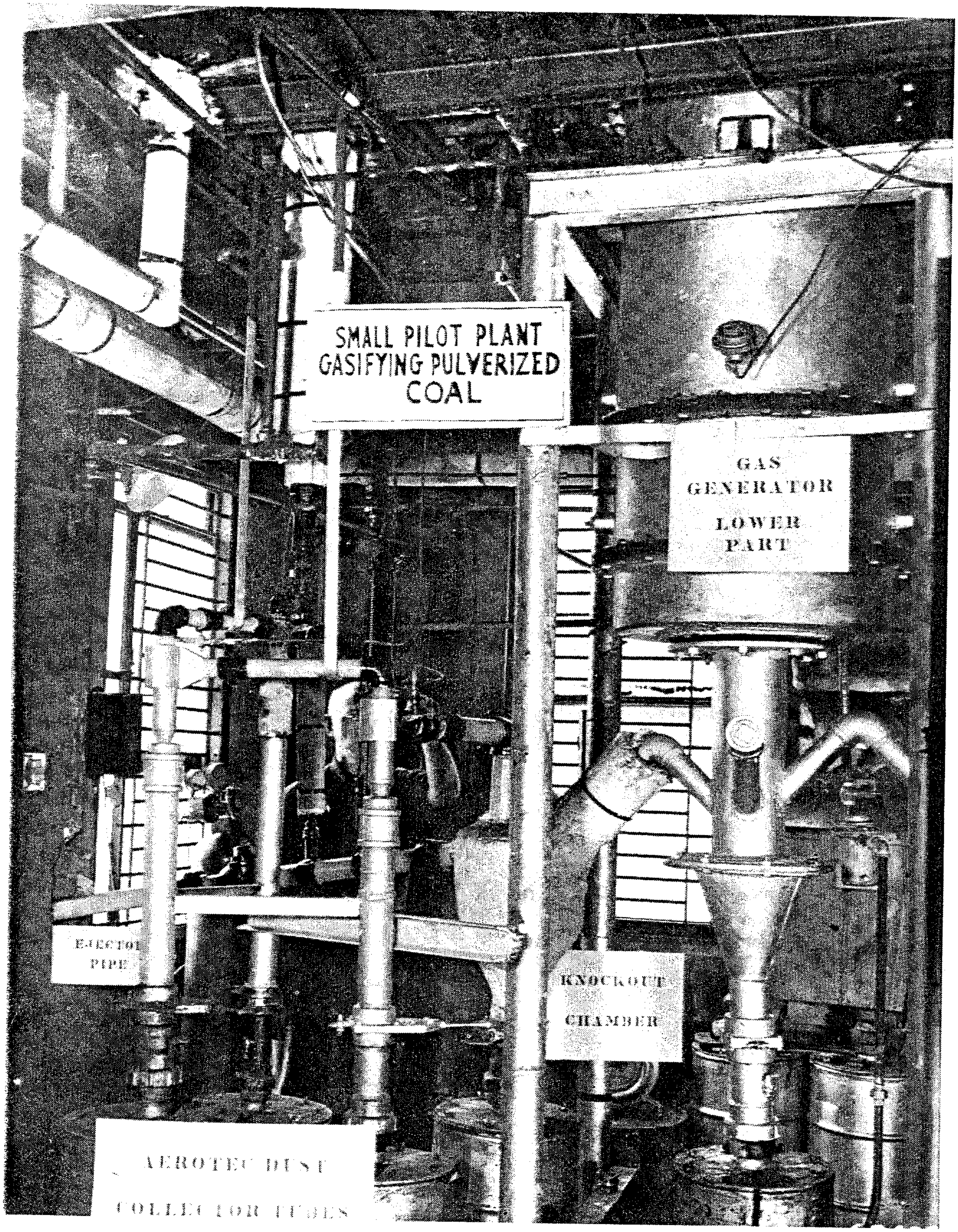

Figure 4. - Laboratory unit (lower floor) in second stage of development.

Figure 1 also shows a steam:oxygen ratio controller called also "proportioner," which served the purpose of saturating the oxygen bubbled through water at given constant temperatures and pressures. Steam could also be admitted through a nozzle in the generator head in case saturation of oxygen did not suffice.

An important part of the generator unit was a water-cooled "head" constructed from alloy-steel (18-8) to prevent its corrosion. Into this 12-inch long, 6-inch I. D. jacketed head, Kemp[16] high-capacity, low-pressure, industrial premix burners were installed at two opposite points for preheating the generator prior to each run with natural-gas - air flames.

The oxygen used in the process was injected, with or without proportioned steam, through two opposing nozzles installed in the generator head at a pressure of 20 to 25 p.s.i.g. From each nozzle a series of oxygen jets were directed downward at such angles (close to 45 degrees) that they impinged upon each other, causing considerable turbulence. Through another pair of nozzles in the generator head natural gas could be introduced for the purpose of supplying very highly superheated steam and CO_2, if desired. In this case, the oxygen introduced was correspondingly increased for complete combustion of the gas.

The powdered coal charged from the Syntron-Feeder fell through the circular opening of a 2-inch, ball-type, Rockwood valve into the gasification chamber. The fine-coal particles, in the form of a dense cloud, reached the impinging oxygen-steam jets, became entrained in the resulting turbulent eddies, and were slowly carried downward by the moving stream of oxygen, steam, and product gases.

Before starting a run, the generator chamber was heated to the necessary reaction temperature, ranging from 2,200° F. at the top to about 1,800° F. at the bottom section. The run was begun by shutting off the preheat burners, turning on the oxygen valve, and starting the feeding of the coal.

A self-explanatory flow diagram in figure 2 shows the gas sampling, testing, and other types of conventional measuring instruments. Actual pictures of the small-scale pilot plant in the second stage of development are shown in figures 3 and 4.

Gasification in improved larger-capacity unit with pneumatic coal-feeding and dry-residue recovery system

The pneumatic coal-feeding system especially developed by the Morgantown Station of the Bureau of Mines[17] permitted charging powdered coal into the generator at a much more constant feed rate and also allowed an increased coal throughput up to about 50 pounds per hour. Charging the coal at a

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[16] Each burner developed 155,000 B.t.u. (gross) per hour, consuming 135 cu. ft. of natural gas per hour.

[17] Albright, C. W., Holden, J. H., Simons, H. P., and Schmidt, L. D., Pneumatic Feeder for Finely Divided Solids: Chem. Eng., vol 56, June 1949, p. 108.