

Coal Preparation and Handling Equipment

The pilot-plant facilities were improved greatly during the year by the installation of enlarged coal preparation and handling facilities. The high-pressure gasifier originally was designed for 6-hour runs. After a series of runs at 300 p.s.i.g. the decision was made to design and install the coal-handling equipment necessary for making runs of 10 hours or longer at 450 p.s.i.g. In this new system run-of-mine coal at the rate of 2,500 pounds per hour is put through a hammer mill and delivered to a standard Raymond mill. The coal is ground to 70 to 90 percent through a 200-mesh screen and, after passing through a 20-mesh scalping screen, is transferred to one of four low-pressure storage tanks. These, with the screw conveyor for filling them, are shown in figure 51. These tanks are equipped for local fluidization of the coal charge. The fluidized coal from these can be transferred by pipeline to high-pressure weigh tanks placed on platform scales (see fig. 52). From these tanks the coal is pneumatically conveyed to the coal feeders at the gasification units. This system has the advantages of pipeline transportation of coal with a minimum of moving parts. It is shown diagrammatically in figure 53. Development work was continued on recycling fluidizing gas for the high-pressure feeder; this gas is apt to contain coal dust. Tests of a lobe-type blower housed in a pressure vessel showed promise, but the best results have been obtained with a high-speed single-stage centrifugal compressor.

Pneumatic Transportation of Coal by Pipeline

Work was initiated on a study of the possibilities of transporting pulverized coal by pipeline. The Morgantown fluidized feeder has been used to transport 500 pounds of pulverized coal per hour through a 60-foot length of pipe of 0.4-inch internal diameter, using only 1 pound of transporting air for over 230 pounds of coal and very low pressure drops along the pipe. It is interesting to note that, under these conditions, approximately 20 pounds of coal per cubic foot of conveying gas or approximately 260,000 B.t.u. per actual cubic foot can be transported. This would compare with approximately 47,000 B.t.u. per cubic foot of natural gas transported at an average pressure of 690 lb. per sq. in. Extrapolation of the data obtained on small-diameter pipes to pipe sizes of interest in commercial transportation of coal shows promise that transportation costs may be very low by this method. Further work is planned on this most interesting project.

Steam-Oxygen Superheater

A combination boiler, superheater, and oxygen preheater was designed and installed in cooperation with the Babcock & Wilcox Co. This equipment will flash 1,000 lb. per hr. of water to steam and superheat it to 1,000° F. and simultaneously preheat 12,000 std. cu. ft. per hr. of oxygen to 1,000° F. This unit has operated very well under very severe conditions. Owing to line and valve losses and to pressure drop through the oxygen preheater and a pressure 450 p.s.i.g. in the gasifier, a line pressure of above 600 p.s.i.g. is required. This necessitates special valves for oxygen service, which have been developed in cooperation with valve companies.

Coal-Preheating Tests

If the coal stream, as well as the oxygen-steam stream, enters the reactor preheated, less time is required for starting the gasification reactions because the preheating need not be done in the reactor. Furthermore, more heat is added to the reactor, decreasing the effect of heat loss - an important item in small-scale experiments. With the nozzle now in use in both gasifiers where the coal stream is broken up by steam-oxygen jets, preheating the coal stream both increases its velocity and decreases its density, making mixing with the gas stream more readily accomplished.

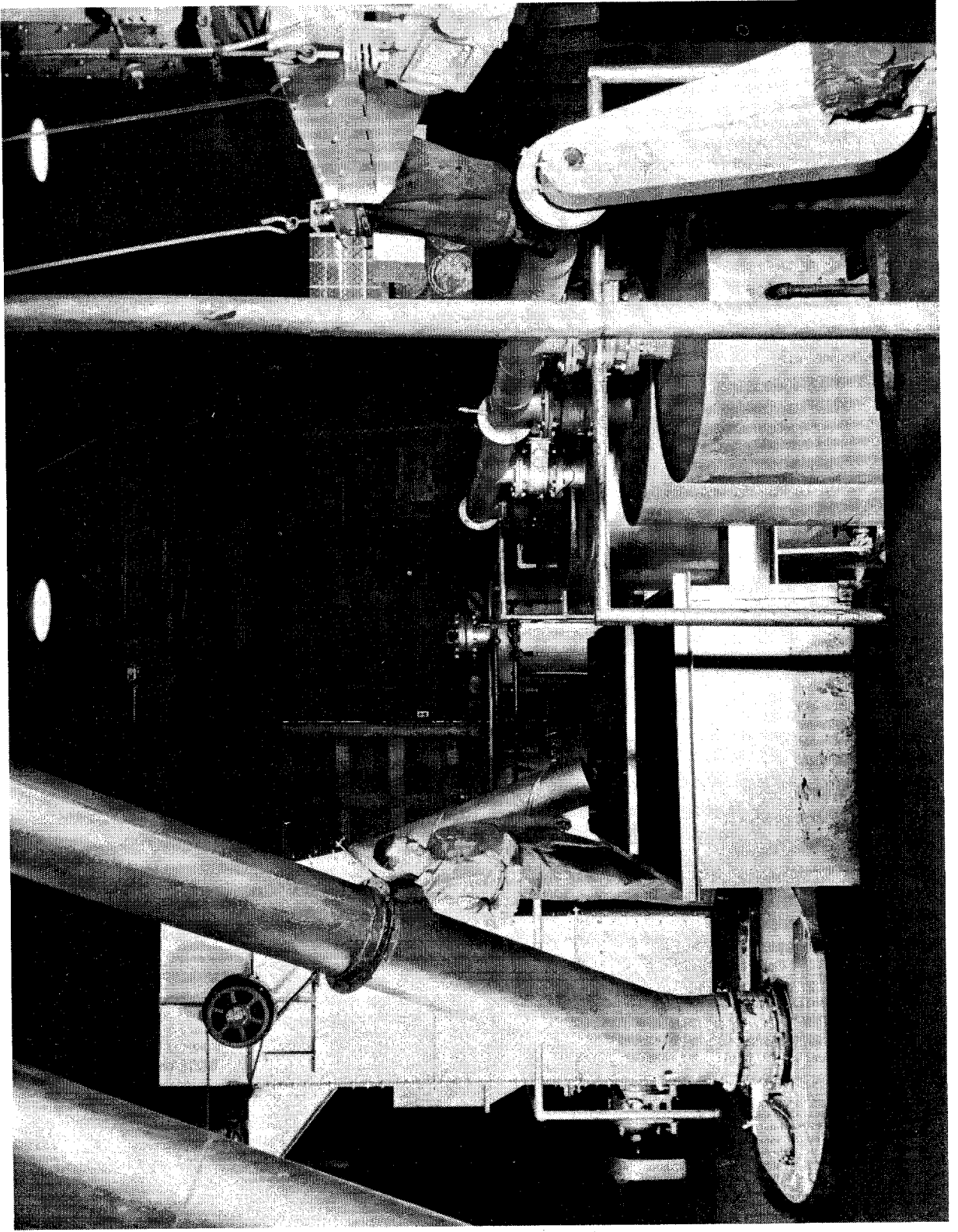


Figure 51. - Coal-preparation system in gasification pilot plants.

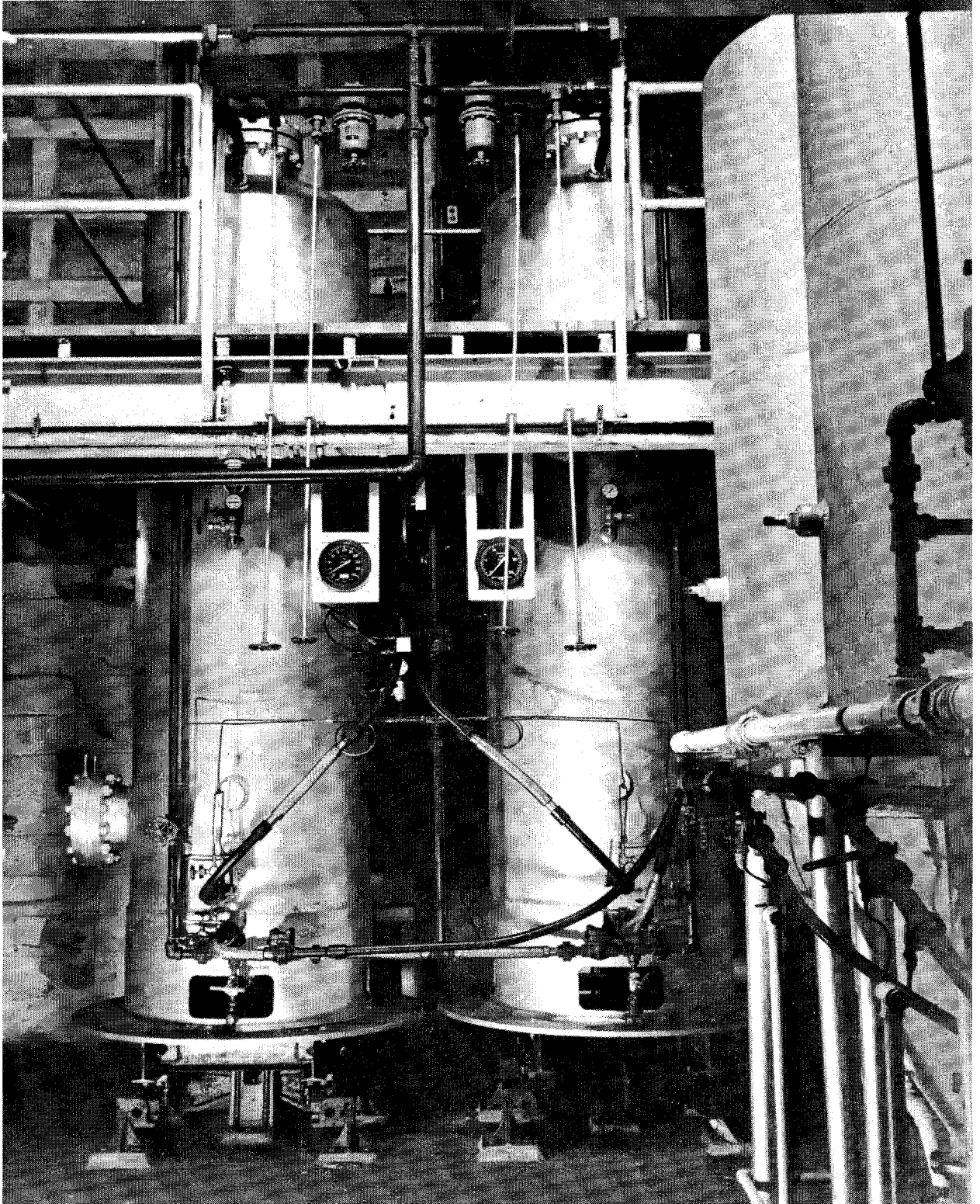


Figure 52. - Coal weigh tanks for pneumatic coal-conveying system.

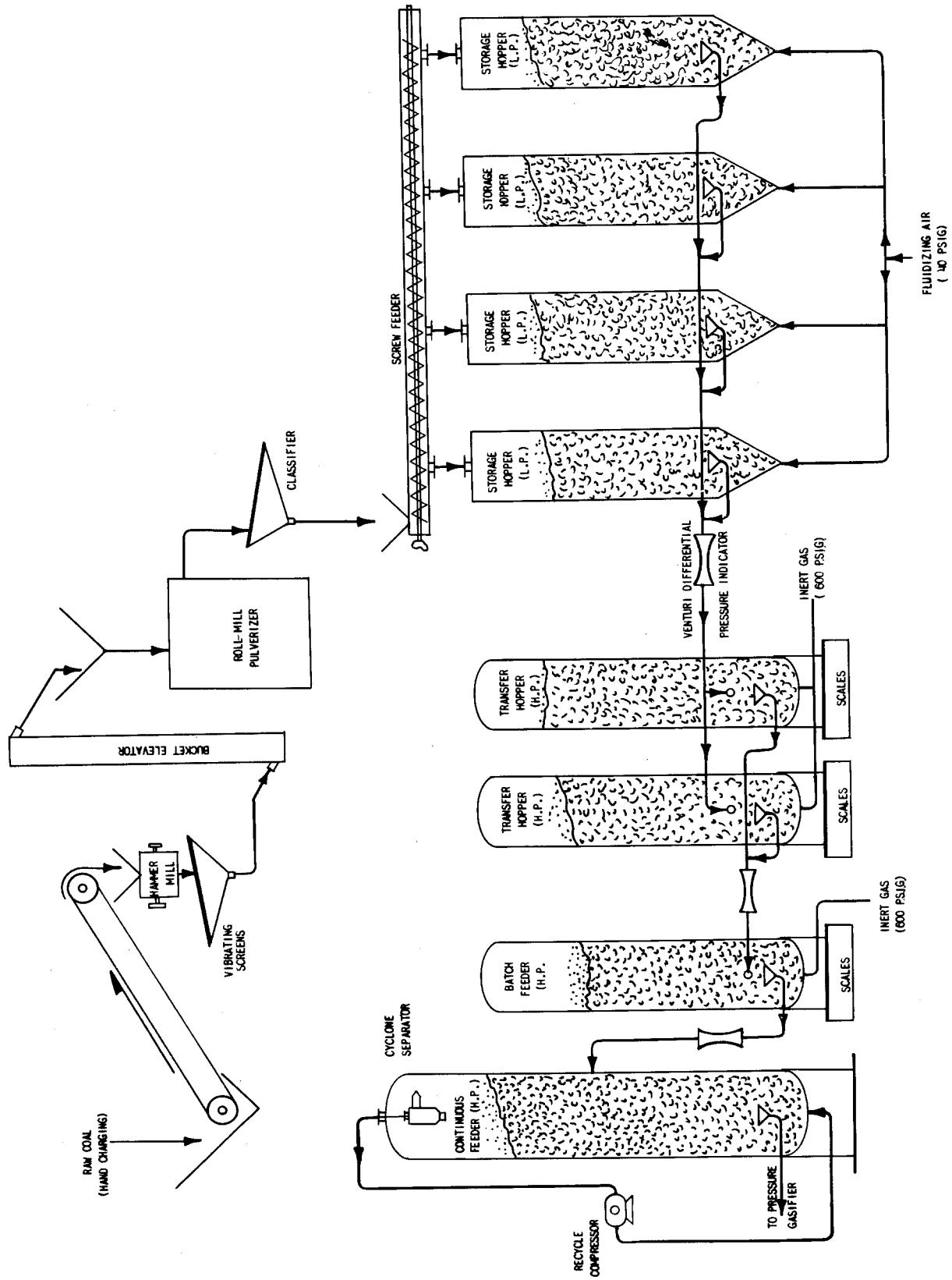


Figure 53. - Coal-handling system for pressure gasification.

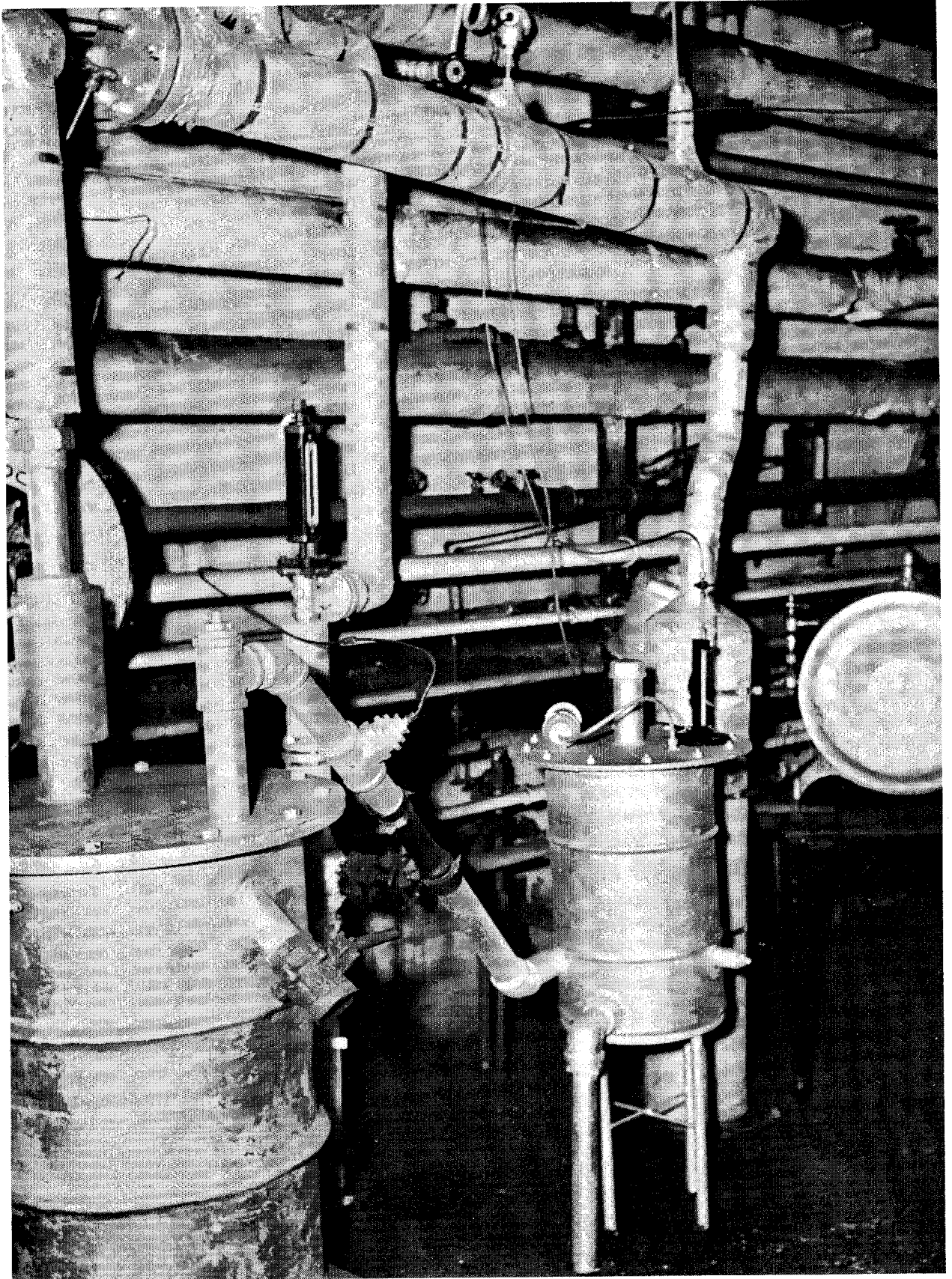


Figure 54. - Dowtherm experimental apparatus for preheating coal.

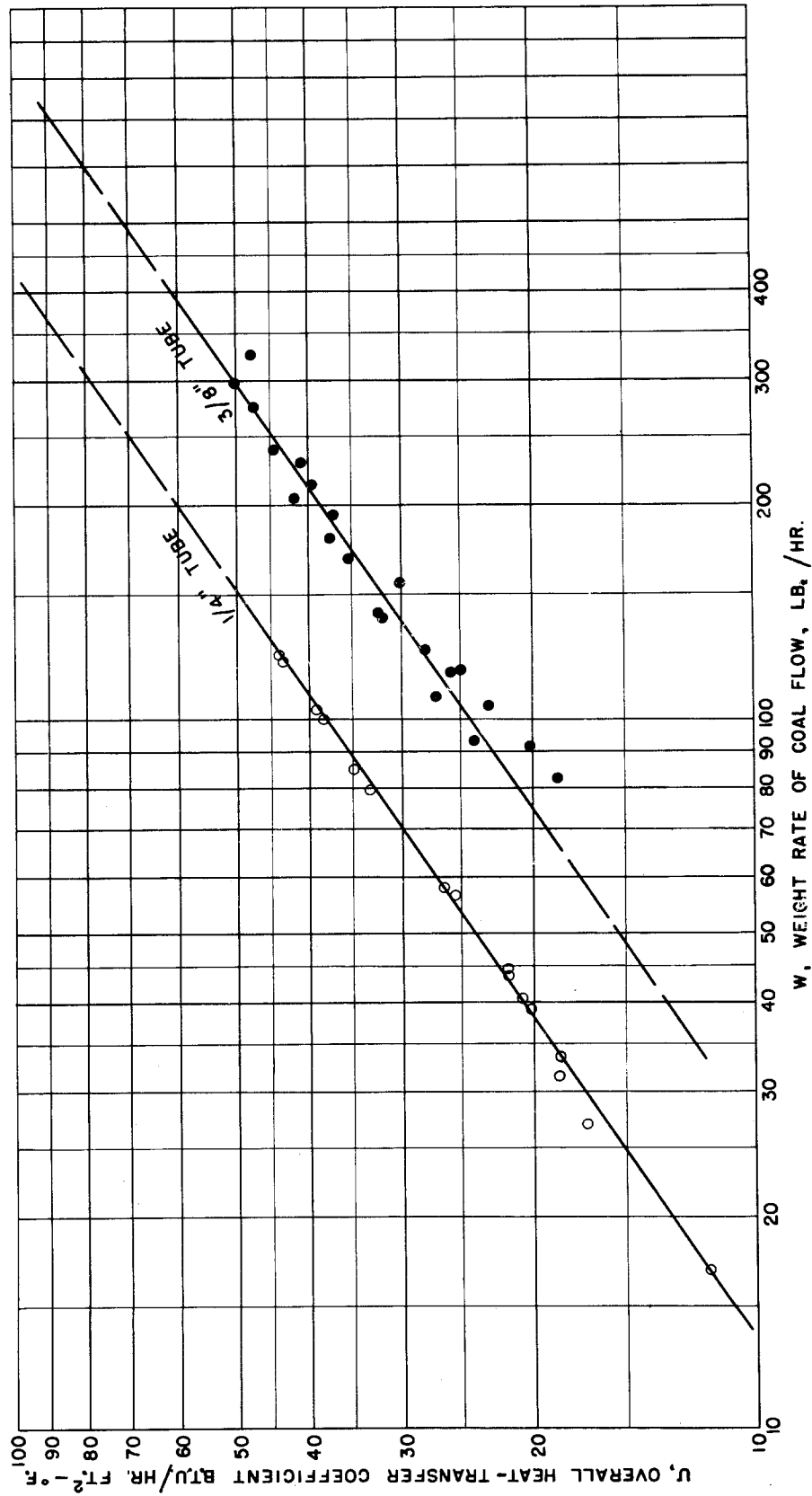


Figure 55. - Effect of weight rate of coal flow on heat-transfer coefficient.

Two sets of experiments have been performed on coal preheating. The objective of the first set was to determine whether coal could be preheated without sticking. The apparatus consisted of a heat exchanger made of a spiral coil of 1/4-inch copper tubing inside a shell of 6-inch pipe. The coal stream from the feeder flowed inside the tube, while hot flue gas from the combustion of natural gas and air in an external furnace flowed outside the coil countercurrently to the coal stream. The flue gas was baffled to increase its velocity and length of path.

Although the exit coal temperature was about 700° F. in most of the tests, Sewickley coal, a strongly coking coal, was heated as high as 900° F., and Rock Springs, Wyo., coal, a very weakly coking coal, was heated as high as 1,100° F. No evidence of any progressive build-up inside the tube was found for a tube that was used for coal heating, for over 40 hours. The tube was plugged three times, but in each instance it is believed that the plugging was caused by a flow stoppage, the coal being coked while not in motion by the high-temperature flue gas used at the time. It is believed that as long as the temperature of the heating medium is below that of the coal's plastic range, no plugging difficulties will occur. The over-all heat-transfer coefficients measured in these tests ranged between 8 and 12 B.t.u. per hr. per sq. ft. per °F. and were essentially equal to the heat-transfer coefficient for the flue-gas film.

The second set of experiments was undertaken to provide data for the design of a heat exchanger to be used in the pilot plant. The apparatus consisted of a 6-foot length of 4-inch pipe inside which was placed a 24-foot section of 1/4-inch steel tubing having three U-bends. The coal stream passed through the tubing, and Dowtherm vapor condensed on the outside of the tube. Figure 54 shows the experimental set-up used in the tests. The data obtained are shown plotted as the effect on the over-all heat-transfer coefficient of the weight rate of coal flow for two tube diameters, namely, 1/4-inch and 3/8-inch O.D. steel tubing (see fig. 55). Since the condensing Dowtherm coefficient was in general greater than 200 B.t.u. per hr. per sq. ft. per °F., the over-all coefficient as plotted essentially equals the coal-stream coefficient. No plugging difficulties were encountered in these tests, since the condensing Dowtherm temperature (650° F.) was below that at which the coal melted. From the data obtained in these tests, a heat exchanger has been designed for preheating the coal stream to about 550 °F. before it enters the gasifier.

Laboratory-scale Experimentation

A program of developments of a small-scale gasifier and of procedure for preliminary testing of powdered solid fuels for their relative value in synthesis-gas production has been completed. The effects of process variables have been analyzed and evaluated; and, the initial objectives of the program having been accomplished, the apparatus has been dismantled. In the tests, powdered coals at 50-pound-per-hour feed rates were gasified in entrainment in oxygen and superheated steam under precisely controlled conditions. The space previously occupied by the gasifier and auxiliary train is now used for other small-scale experiments pertinent to coal gasification.

Process Design and Cost Studies

Preliminary process calculations have been made, and corresponding tentative flow sheets have been prepared for a 150-ton-per-day ammonia plant using the Bureau of Mines powdered-coal-gasification process to produce the synthesis gas required. Although this cost study was carried out under a cooperative agreement with a private firm, with other benefits accruing to the Bureau of Mines, the primary objective has been to obtain guidance in directing development of the coal-gasification process.

A study also was made of the effect of refractory life in commercial-size gas generators on the cost of synthesis gas. Several diagrams were prepared to show the effect of "down time" on the total cost of silicon carbide relinings, including the fixed charges on the gasifier and accessories accrued during down time. The diagrams illustrate the relation between the cost of a unit volume of synthesis gas and number of relinings per year, expressed with relation to percentage of down time. For silicon carbide it was concluded that, if the down time rises above 30 percent of the total time (down time plus operating time) the effect on the cost of synthesis gas may be prohibitive. With 10 percent down time the cost of synthesis gas per 1,000 cu. ft. of $\text{CO} + \text{H}_2$ increases by 0.3 cent, and for 20, 30, and 40 percent down times the cost increases are 0.65, 1.10, and 1.75 cents, respectively.

An estimate also was made of the cost of compressing synthesis gas made at atmospheric pressure for a synthine plant producing 10,000 barrels of liquid fuels per calendar day. This cost estimate was based on compression of 16,800,000 std. cu. ft. per hr. of raw synthesis gas (after dust removal but before sulfur purification and shift conversion) to 450 p.s.i.g. pressure. The cost of investment in 12 turbines, 24 centrifugal 4-stage compressors with intercoolers, water pumps, buildings, and accessory installations was found to be \$14,035,000 as of August 1950. The operating cost, which includes various taxes, insurance, and amortization but no Federal taxes or interest on capital invested, was 5.24 cents per 1,000 std. cu. ft. of synthesis gas compressed.

Gas Purification

Analytical Methods

A method has been developed for semiquantitative determination of ethyl mercaptan in synthesis gas. The method consists of absorbing the mercaptan in caustic, followed by development of a pink color with p-aminodimethylaniline sulfate. The data do not obey the Lambert-Beer law, and the effect of variables on the method is marked. Hydrogen sulfide interferes considerably and must be removed from the gas stream. Carbonyl sulfide (carbon oxysulfide or COS) gives slightly high readings owing to formation of methylene blue.

A reliable gravimetric method for determining moisture in gas was developed. Results from determinations on the make gas from the atmospheric-pressure gasifier were in good agreement with those calculated from material balances using the test-run data.

A method was developed for determining the weight concentration of dust in highly purified gas or atmospheric air by use of a large-area filter paper that can be weighed on an analytical balance. If the flow of gas does not exceed about 5,000 cu. ft. per hr., this apparatus can filter the entire gas stream, eliminating the difficult and inaccurate sampling step.

Bench-Scale Experiments

A limited number of bench-scale experiments were carried out to study the possibility of recovering the hydrogen sulfide as elemental sulfur from an acid gas stream containing only 2 to 3 percent hydrogen sulfide. Such operation would permit substantially complete removal of the hydrogen sulfide and carbon dioxide from raw synthesis gas simultaneously. In these experiments the hydrogen sulfide was oxidized to sulfur, using an iron sulfide catalyst. With an acid gas containing about 2 to 3 percent hydrogen sulfide, a conversion of 78 percent was obtained with one pass over the catalyst. With better control over the temperature and oxygen addition, this conversion efficiency would probably be improved.

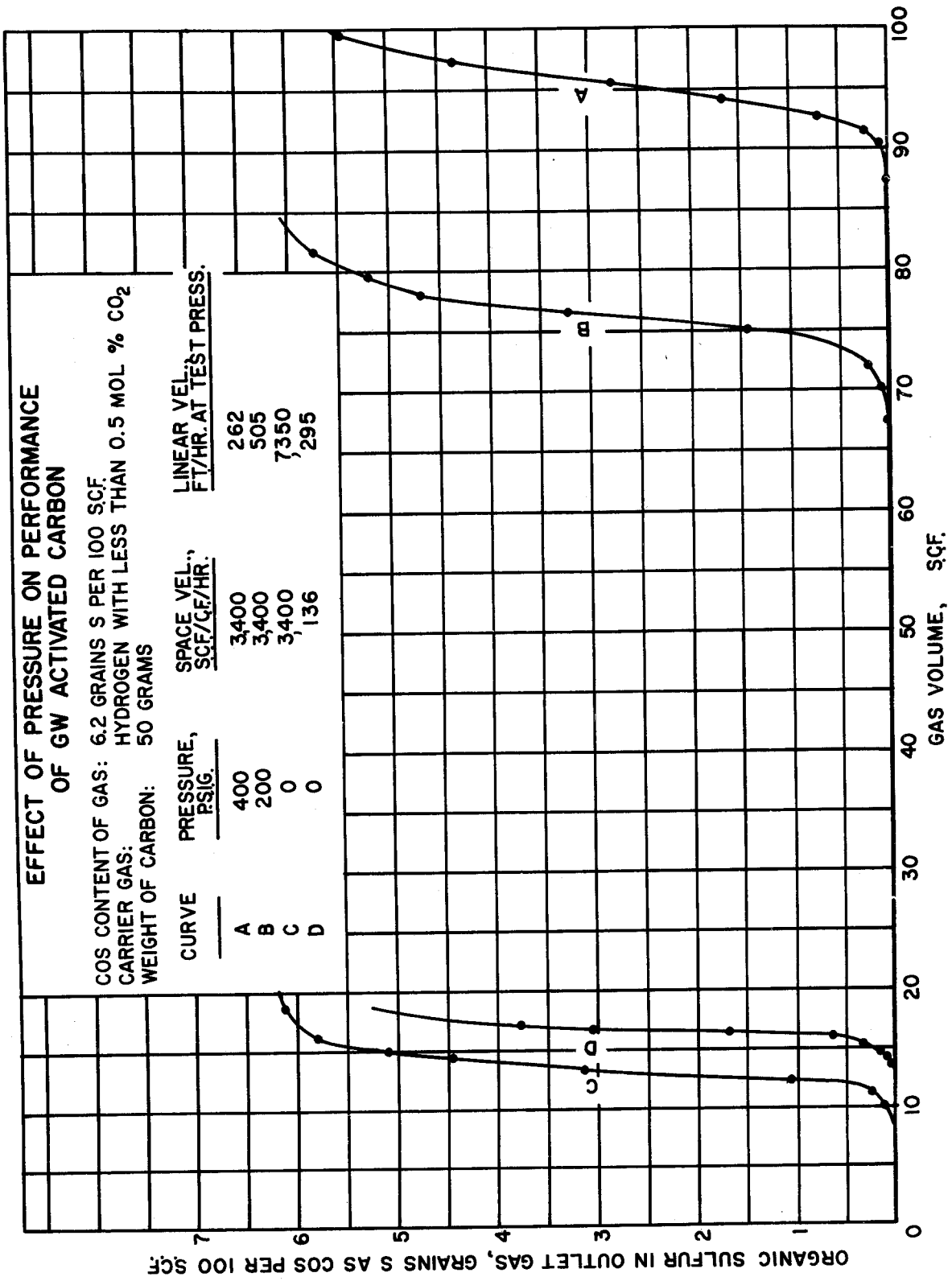


Figure 56. - Effect of pressure on performance of GW-activated carbon.

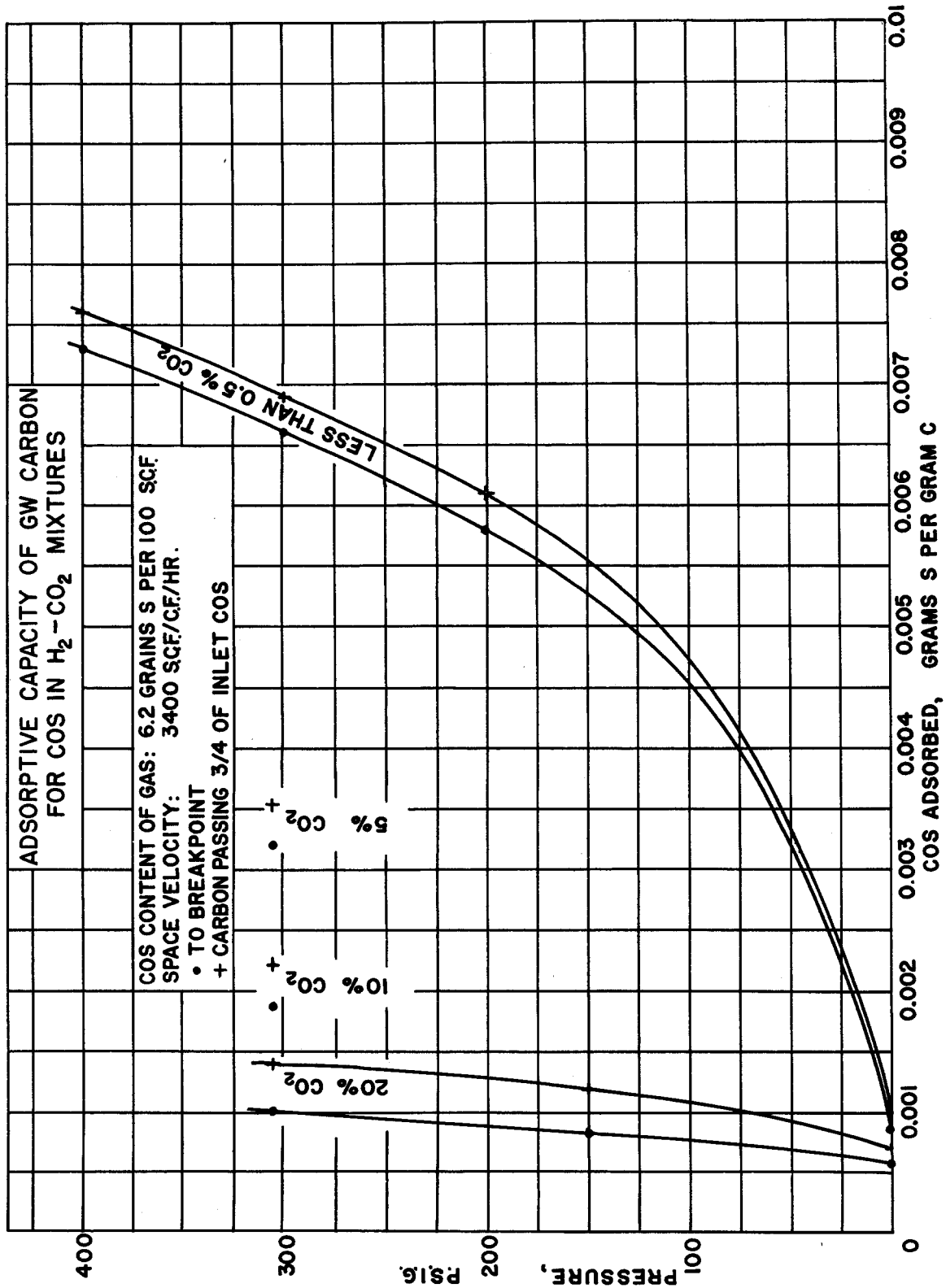


Figure 57. - Adsorptive capacity of GW carbon for carbonyl sulfide (COS) in H₂ - CO₂ mixtures.

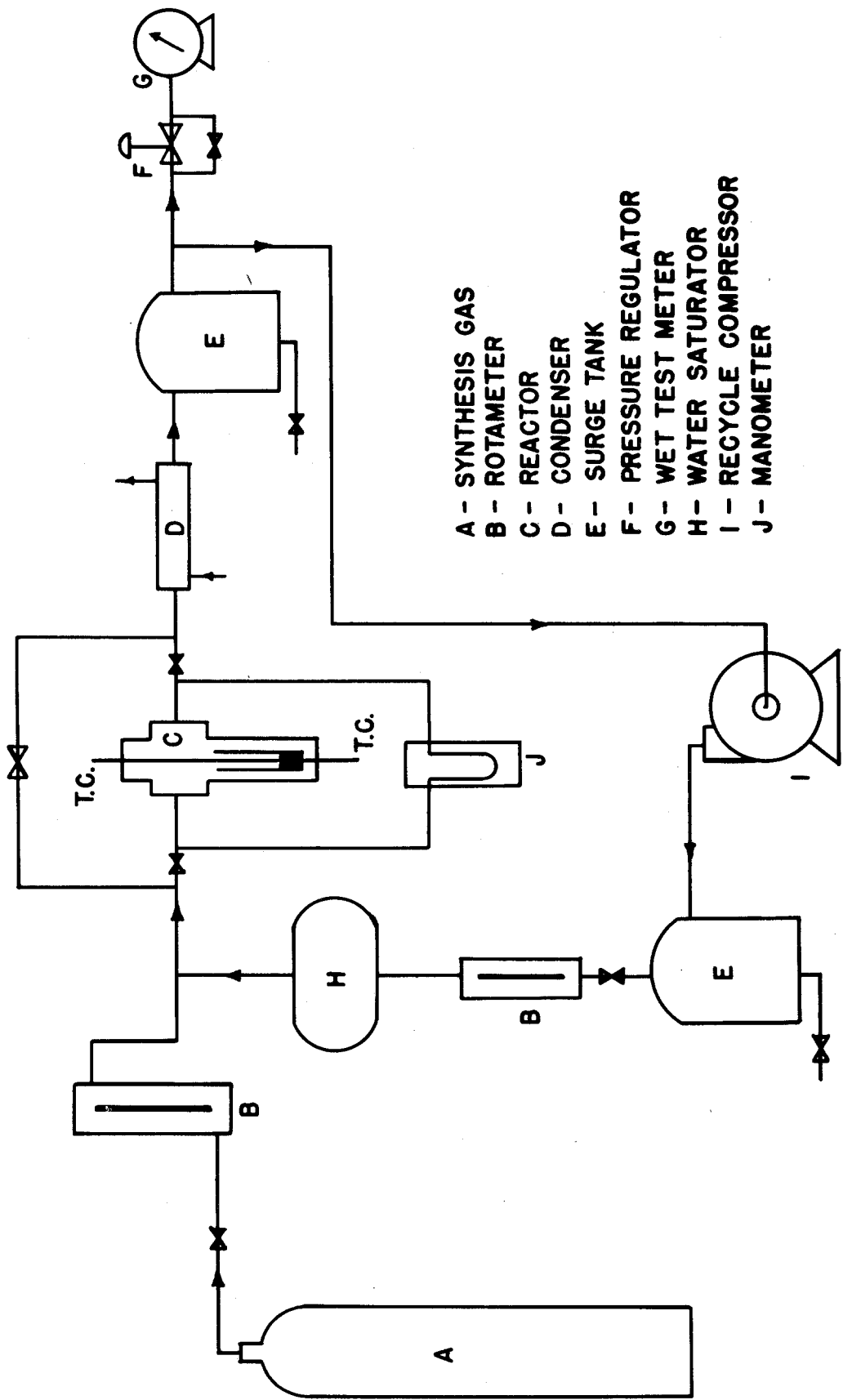


Figure 58. - Diagrammatic arrangement of apparatus for studying synthesis of methane.

Previous experimental work on the effect of pressure on the adsorption of carbonyl sulfide by GW activated carbon showed that about 1.7 times as much sulfide was adsorbed at 300 p.s.i.g. as at atmospheric pressure. This comparison was obtained from bench-scale experiments in which the carrier gas was hydrogen containing 20 percent carbon dioxide. In actual plant operation, however, the carbon dioxide content of the gas entering the activated-carbon purifiers is considerably under 20 percent. Therefore, bench-scale tests were made to study the effect of pressure on removing carbonyl sulfide from gas containing low amounts of carbon dioxide.

Hydrogen containing 6.2 grains of sulfur as carbonyl sulfide per 100 std. cu. ft. and less than 0.5 mol. percent, carbon dioxide was used in the tests. After each test, the carbon was regenerated by passing nitrogen through the bed 1 hour at 500° F. Data from four runs are plotted in figure 56. During runs A, B, and C the space velocity rather than the linear velocity was held constant. This resulted in a linear velocity of 7,350 ft. per hr. for the run at atmospheric pressure, about 30 times that used in the run at 400 p.s.i.g. To make certain that the difference in adsorptive capacity was not due entirely to the difference in linear velocities, run D was made at atmospheric pressure, using a linear velocity corresponding to that at 400 p.s.i.g.

Figure 57 shows the effect of pressure on the adsorptive capacity of GW carbon for carbonyl sulfide when operating with gases of different carbon dioxide content. Capacities are given to the point where the carbon begins passing the first trace of sulfur and to the point where it is passing three-fourths of the inlet concentration. It can be noted that, when only a small amount of carbon dioxide is present in the gas, the carbon adsorbs about 7.5 times as much carbonyl sulfide at 300 p.s.i.g. as at atmospheric pressure. As the carbon dioxide content of the gas increases, the difference in the capacity at the two pressures decreases until, with a gas containing 20 percent carbon dioxide, the carbon adsorbs only 1.7 times as much at 300 p.s.i.g. as at atmospheric pressure.

As part of a program for investigating the practicability and cost of producing pipeline gas from coal, a bench-scale experiment was carried out under a cooperative agreement between the Bureau of Mines and the Southern Natural Gas Co. on the conversion of synthesis gas to methane. The objective of the experiment was to determine the economic factors involved in the methane-synthesis step, using a catalyst that had already been developed for such a process. The Bureau was especially interested in such a program, as it afforded an opportunity of studying the applicability and efficiency of its gas-purification system in rendering synthesis gas suitable for methanation processes. As the purification system developed at Morgantown can supply a purified gas containing no more than 0.05 grain of sulfur per 100 std. cu. ft., the methanation experiment was carried out using simulated synthesis gas containing from 0.02 to 0.05 grain of sulfur per 100 std. cu. ft.

Figure 58 gives a diagrammatic arrangement of the apparatus used in the experiment. The experiment was made at a pressure of 300 p.s.i.g. and a fresh synthesis-gas flow equivalent to a space velocity of 2,000 std. cu. ft. per cu. ft. per hr. As the synthesis reaction is strongly exothermic, temperature control was maintained by recycling part of the product gas. The recycle gas, before mixture with the fresh gas, was bubbled through a water saturator with the temperature so controlled to give a gas containing from 18 to 20 percent steam. The recycle - gas-fresh-feed-gas ratio was maintained at 12:1. The results from the one experiment were inconclusive and future experiments are being planned.

Operation of Pilot Plant

As part of a development program for investigating means of removing dust from synthesis gas, pilot-plant runs have been made using a moving-bed filter. Coke was

used as the filtering medium. When used with a simulated dust-laden gas, the efficiency of removal varied from 94 to 99.98 percent, depending principally on the size of dust particles or agglomerates in the gas stream. When the filter was operated as part of the purification train of the atmospheric-pressure gasifier, efficiencies ranging from 94 to 99.3 percent were attained. Data indicated that the pressure drop across the coke bed, resulting from build-up of dust on the coke, had a considerable effect on the removal efficiency. An increase in the pressure drop resulted in higher efficiencies.

Pilot-plant runs were continued throughout the year on removal of organic sulfur by activated carbon and simultaneous removal of hydrogen sulfide and organic sulfur using a copper-chromium-vanadium catalyst. Figure 59 shows the pilot-plant equipment used in the catalytic studies. The preliminary runs indicated that the pilot-plant results corroborated those obtained from bench-scale experiments.

One run has been made using two reactors in series, each containing 0.3 cu. ft. (26.4 pounds) of catalyst. The run was made at a pressure of 90 p.s.i.g. and space velocities up to 5,000 std. cu. ft. per cu. ft. per hr. Temperatures in the first reactor ranged from 760 to 820° F. and in the second from 570 to 630° F. The raw synthesis gas contained 425 grains of hydrogen sulfide and 31.5 grains of organic sulfur per 100 std. cu. ft. Until the break point, the gas leaving the first reactor contained less than 0.01 grain of sulfur as hydrogen sulfide and less than 0.10 grain of sulfur as organic sulfur. The gas leaving the second reactor always contained less than 0.01 grain of total sulfur per 100 std. cu. ft. At the break point, the catalyst in the first reactor has absorbed 11 percent of its weight in sulfur.

Earlier pilot-plant investigations dealt with removal of carbon dioxide and hydrogen sulfide with di- and tri-ethanolamine. During the past year, liquid-purification studies were continued on selective removal of hydrogen sulfide from gas containing relatively large quantities of carbon dioxide. Aqueous solutions of tripotassium phosphate and Alkacid DIK were investigated. All purification studies in the pilot plant were carried out at 300 p.s.i.g., the approximate pressure contemplated for the Fischer-Tropsch synthesis. Most of the pilot-plant runs were made using inert gas obtained by burning natural gas. Hydrogen sulfide and additional carbon dioxide, if necessary, were added to the inert gas to give the desired concentration. Synthesis gas was used when available.

Results from the Morgantwon pilot-plant operation showed that, when a gas having a carbon dioxide - hydrogen sulfide ratio of 15:1 was treated, the phosphate solution absorbed about 0.03 pound of sulfur per gallon of circulating solution and the Alkacid, about 0.04 pound per gallon. However, the selective properties of the phosphate solution appeared to be better than those of the Alkacid. With a 15:1 ratio in the untreated gas, the phosphate process gave an acid-gas (carbon dioxide and hydrogen sulfide) mixture leaving the reactivator that contained about 23 percent hydrogen sulfide. When such a gas was treated with the Alkacid DIK solution, the acid gas contained about 14 percent hydrogen sulfide.

Neither the phosphate nor the Alkacid solutions affected, to any great extent, the organic sulfur content of the synthesis gas. Analyses showed that the little that was removed by the phosphate did not build up in the lean solution. It is believed that part of the carbonyl sulfide may have been hydrolyzed and removed as potassium bisulfide.

Proposed Pressure Gasifier on Pilot-Plant Scale

Preliminary designs are being worked up for a pressure gasifier complete with waste-heat boiler and slag-removal system planned for installation in the new Morgantown Experiment Station. To make this unit as flexible as possible, the design is

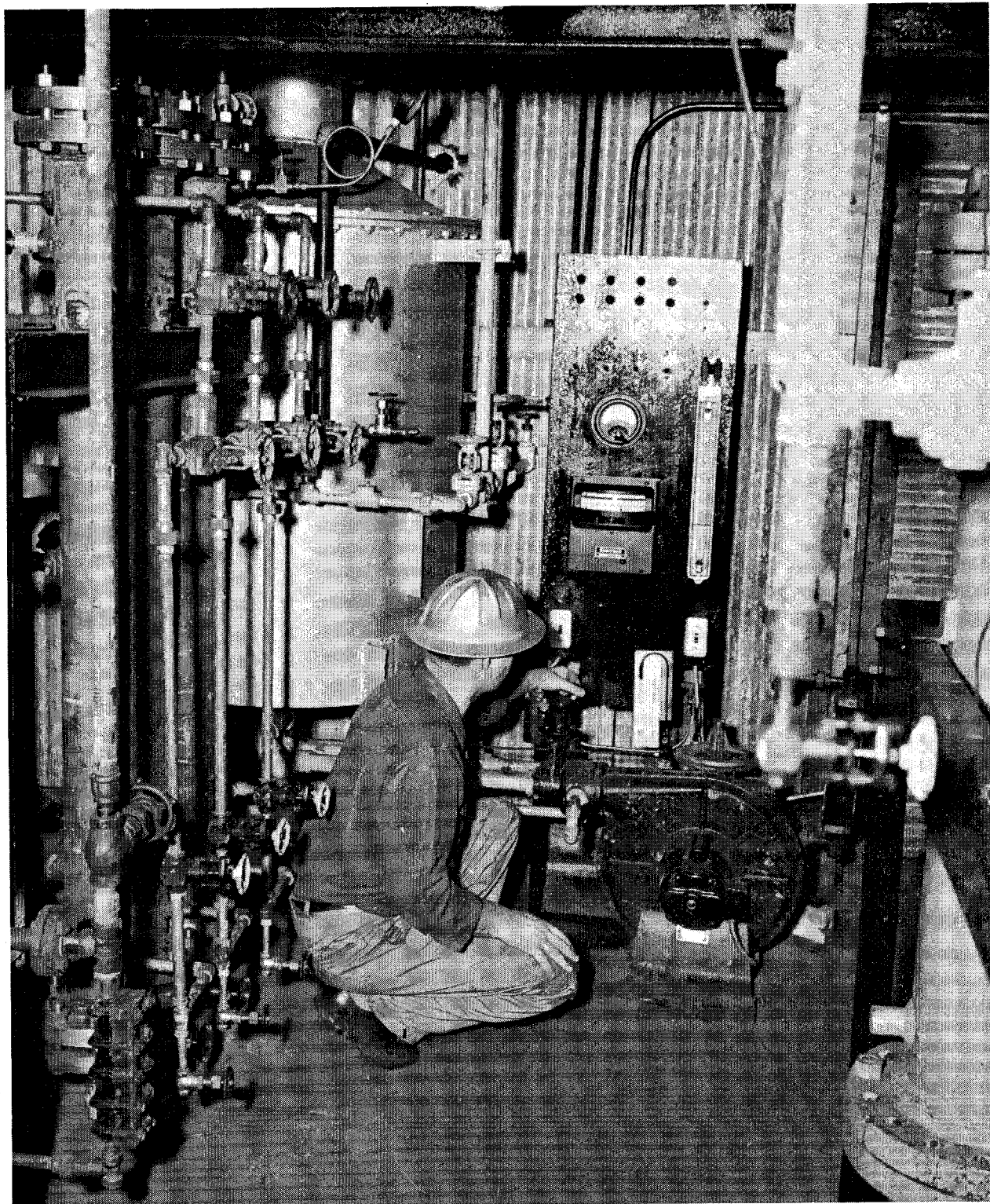


Figure 59. - Pilot-plant apparatus for removing hydrogen sulfide and organic sulfur compounds from gas simultaneously.

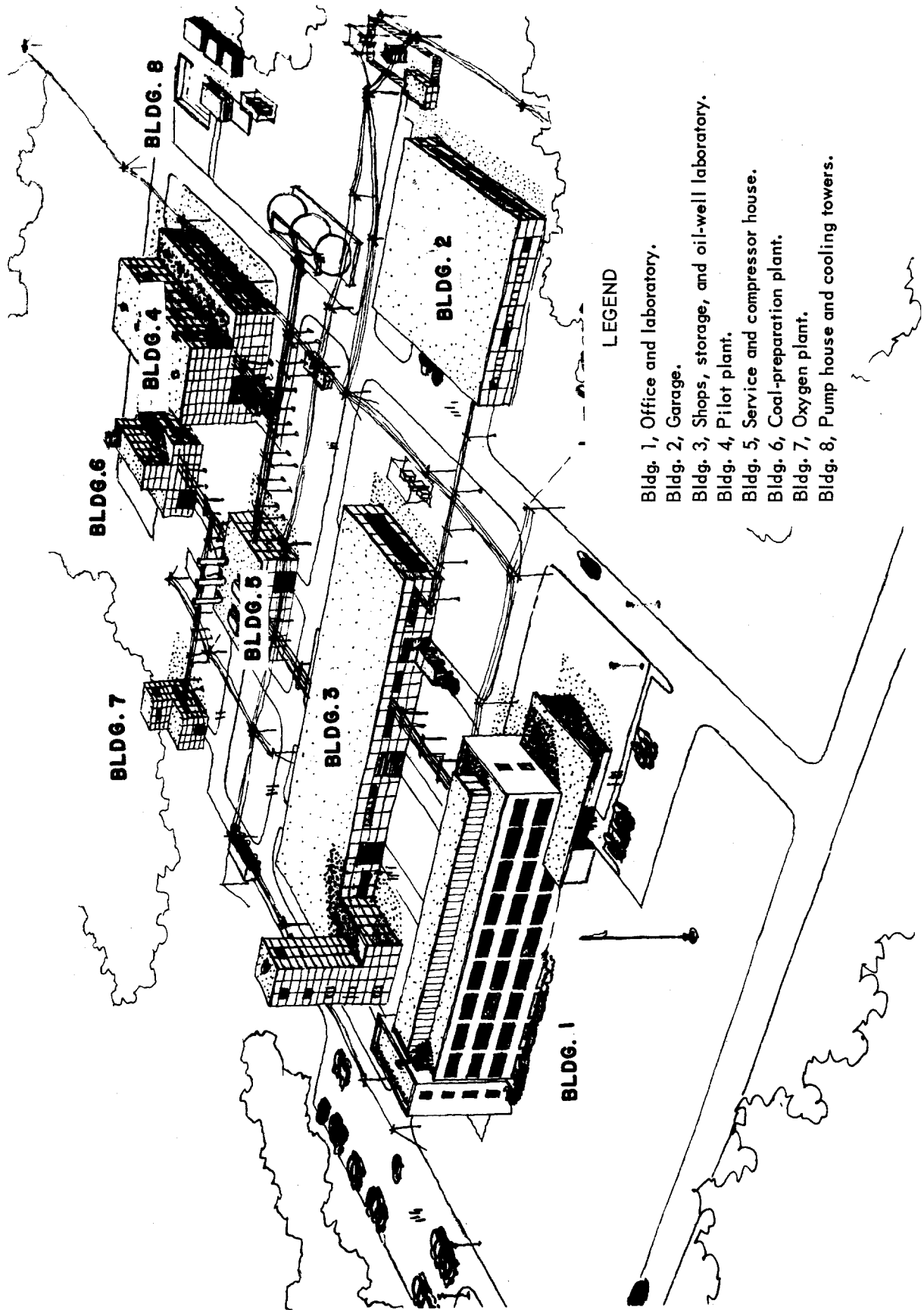


Figure 60. - Architect's sketch of new Morgantown, W. Va., Experiment Station.