Gas Purification

Raw gas produced from coal contains dust and gaseous impurities, most of which must be removed before the gas is used. Work has been done on the determination and removal of dust particles and on desulfurization.

Analytical Methods

Suitable methods had been developed previously for determining the concentration of dust and moisture in crude or highly purified synthesis gas. Attention has been concentrated recently on particle-size determination. An absolute method is being developed for determining particle-size distribution in dust by direct microscopic observation. First, the dry dust is dispersed in a suitable liquid to break up agglomerates. An aliquot portion of the slurry is then placed in a flat dish, the particles are allowed to settle, and the liquid is evaporated without movement of the particles. The method has been improved by use of optimum dust concentrations in the slurry, by slow drying, and by control of the drying air to prevent drafts. Particle size is measured with a calibrated grid in the microscope eyepiece.

Although tedious, this method gives absolute values that can be used to standardize more rapid methods. Also, microscopic observation gives information about particle shape.

This method has been improved so that it is reasonably satisfactory for medium and large particles. With finer particles dispersion of agglomerates was incomplete. It was improved in liquids of high viscosity. The poor results with fine particles were due in part to surface-tension movement of the particles during final drying. These problems will assume still greater importance as even finer particles are investigated.

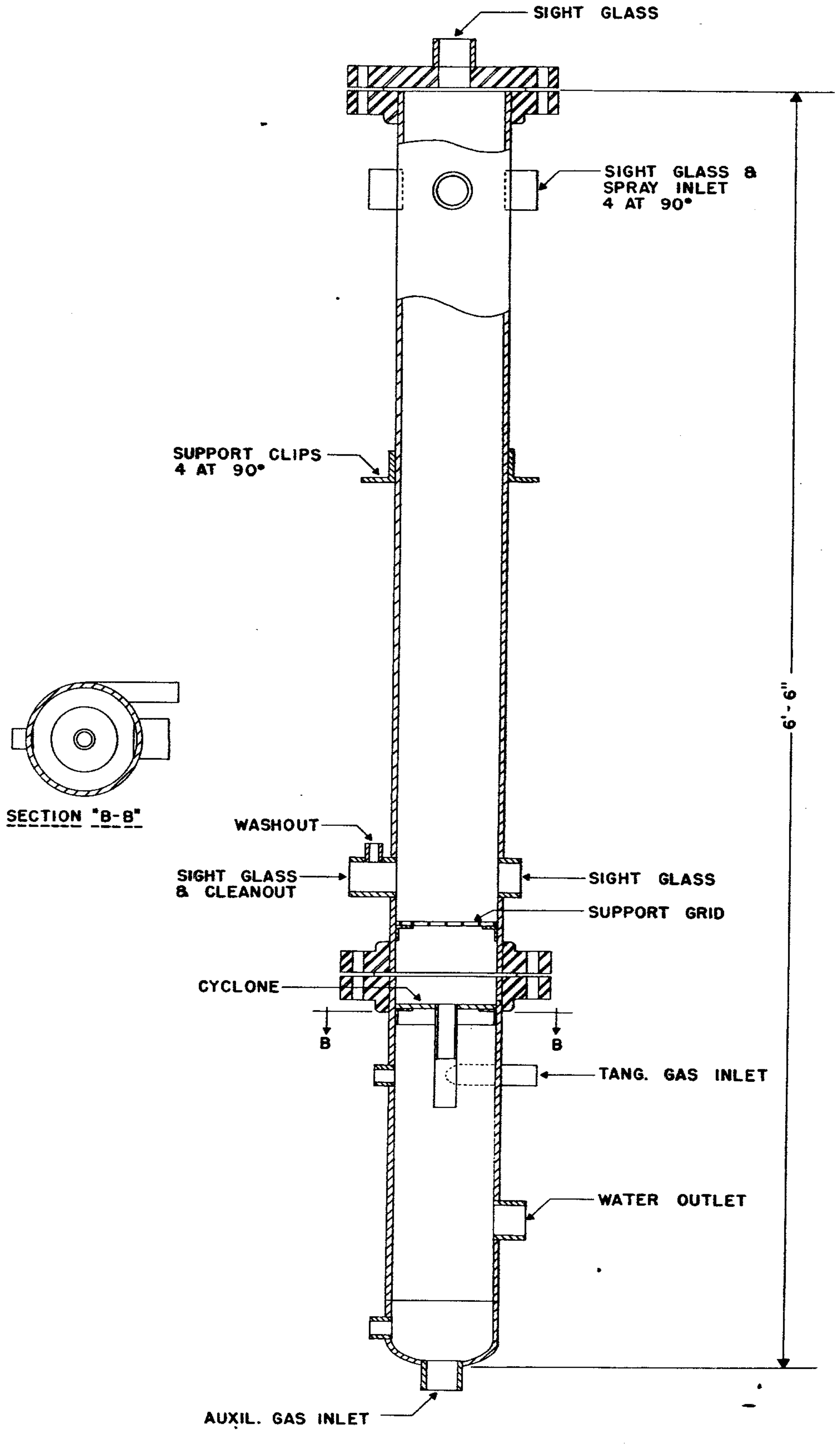
More important than ultimate particle size of a dust is the size of particles as present, in more or less agglomerated condition, in a gas stream. Much of the available information about dust-removing equipment is of little value because the size distribution of dust particles in the gas stream is unknown.

The membrane filter was tried for determining actual particle size. The filter is really a sieve with holes 0.8 micron in diameter rather than a true filter with a deep mat. When gas is passed through, particles remain on a flat surface and can be readily counted and examined under a microscope.

A source of trouble was static electricity, which caused particles to jump from the plastic filter material, especially under dry conditions; much less trouble was encountered in summer. Attempts will be made to extend the range of usefulness of these filters. At present the method is suitable only for highly purified gases because only a small amount of dust can be handled on the filter.

Bench-Scale Experiments

Tests are being made of dust-removing equipment used in the pilot-plant gasification systems, such as scrubbers, electrostatic precipitators, and moving-bed filters. High-pressure scrubbing in a tower packed with Raschig rings had proved very effective on a pilot-plant scale, but conditions could not be controlled to permit determination of the effects of variables, such as dust size, dust concentration, gas flow, and water flow. For this reason, a laboratory scrubber (see fig. 9) has been constructed. Below the ring-packed tower is a cyclonic section that can be removed, if desired.



TEST SCRUBBER
D-3398

Figure 9. - Scrubbing tower.

Testing the scrubber or other laboratory-scale dust-removing devices requires a method for producing a dust-laden gas stream. The counterflow feeder (fig. 10) is being developed for this purpose. A small fraction of a pulsating gas stream is passed upward through an orifice, counter to the downward flow of dust into the gas stream that passes below. The dust flow can be decreased by increasing the upward flow of gas, which can be measured by determining the pressure drop across the orifice.

Results are highly reproducible. Deviations from the mean dust concentration average only 5 percent; however, careful mixing of the dust before placing it in the feed hopper is necessary to obtain this precision. Proper operation requires some large particles in the dust, but these can be removed with an elutriator. A wide range of dust flow can be obtained with a single orifice.

In connection with desulfurization of gas, aluminosilicates ("molecular sieves") were evaluated as to their effectiveness in removing carbonyl sulfide and hydrogen sulfide from H_2-N_2 and H_2-N_2-CO_2 mixtures. Of those tested, the most effective type adsorbed 0.011 gram of sulfur (as carbonyl sulfide) per gram of adsorbent at 300 p.s.i.g. This capacity, however, is considerably below that of GW activated carbon (0.019 gram of sulfur per gram of carbon). When 5.2 percent carbon dioxide was added to the carrier gas, all the aluminosilicates tested began passing carbonyl sulfide almost immediately.

At atmospheric pressure the best aluminosilicate adsorbed approximately 2 to 3 grams of sulfur as hydrogen sulfide per 100 grams of adsorbent. However, after one regeneration this capacity was reduced by one-half, and additional regeneration resulted in further loss of capacity. Under the conditions of the tests this type had little capacity for removing hydrogen sulfide from an H_2S-CO_2 mixture.

Nuclear Heat for Coal Gasification

In collaboration with the Reactor Development Division of the Atomic Energy Commission, coal gasification by nuclear heat is being studied. Nuclear energy would take the place of energy released by partial combustion of coal. The heat necessary for the reaction between steam and coal would be supplied by uranium-235 or some other nuclear fuel enclosed in suitable containers.

Several designs for a large-scale nuclear gasifier have been considered. In the design preferred at present, the reactor would be a water-jacketed pressure vessel, packed with spheres containing fissionable material enclosed in a refractory coating. This design would provide a large surface to insure a safe rate of heat transfer through the coating. For satisfactory performance, the slag running through the voids between the balls should cause minimum erosion.

To determine the conditions necessary to keep the slag fluid and to study the rate of erosion of fuel elements, an electrically heated bench-scale test reactor will be constructed (see fig. 11). This laboratory-size reactor has been designed to contain 3/4-inch silicon carbide balls inside a 26-inch-long, 2-inch-I. D. silicon carbide tube, which will be electrically heated by a carbon-spiral resistor. Steam and coal will be passed downward through the bed. The conditions will simulate on a small scale those in large reactors where heat for the steam-carbon reaction, at temperatures up to 3,000° F., may be provided by radioactive material enclosed in refractory shells. This laboratory-scale reactor will serve chiefly to test slag and coal erosion of silicon carbide and other materials, heat-transfer limitations, and temperature resistance of materials. In addition, the apparatus

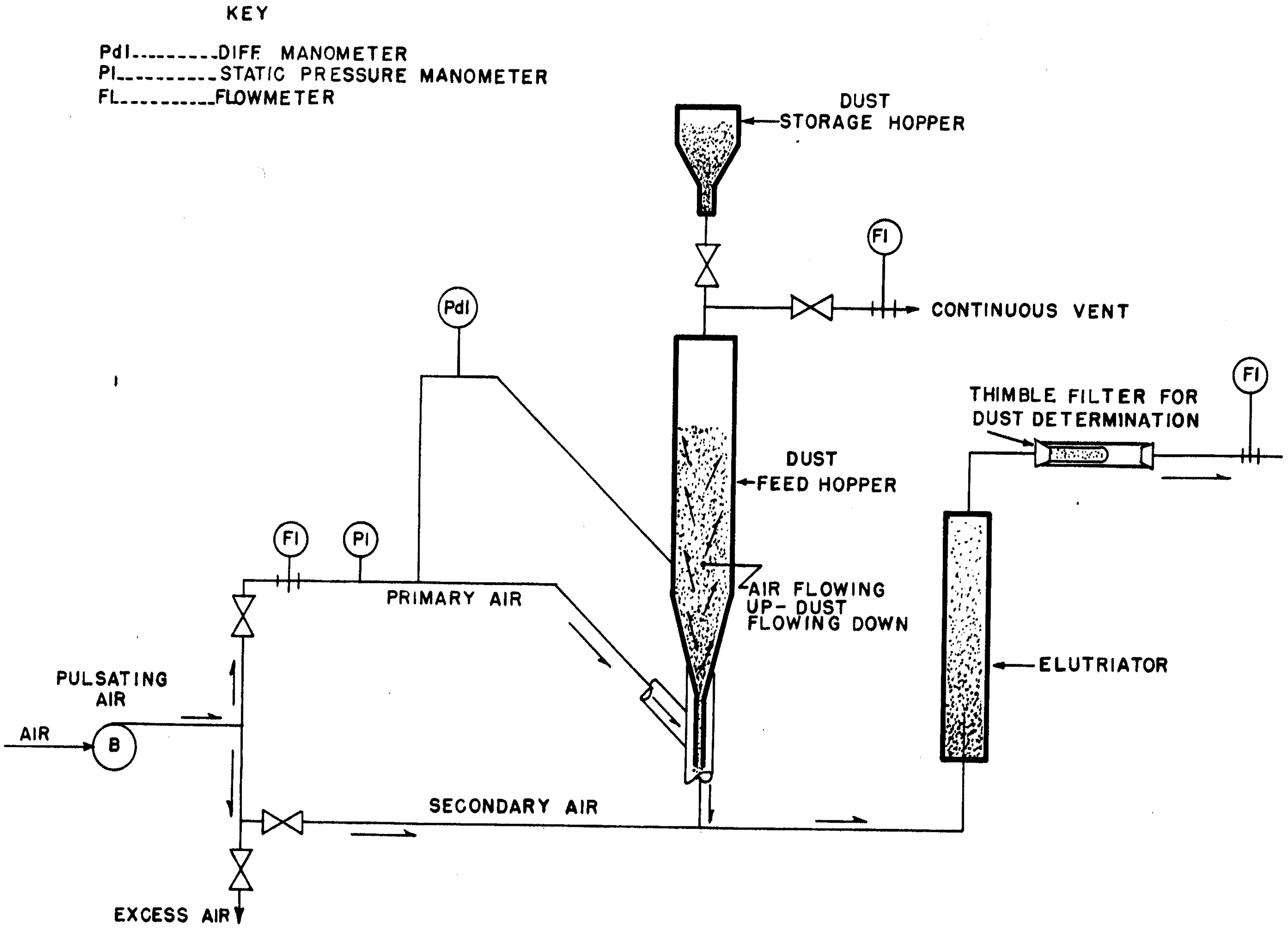


Figure 10. - Flow diagram of laboratory-scale pulsating counterflow dust feeder.

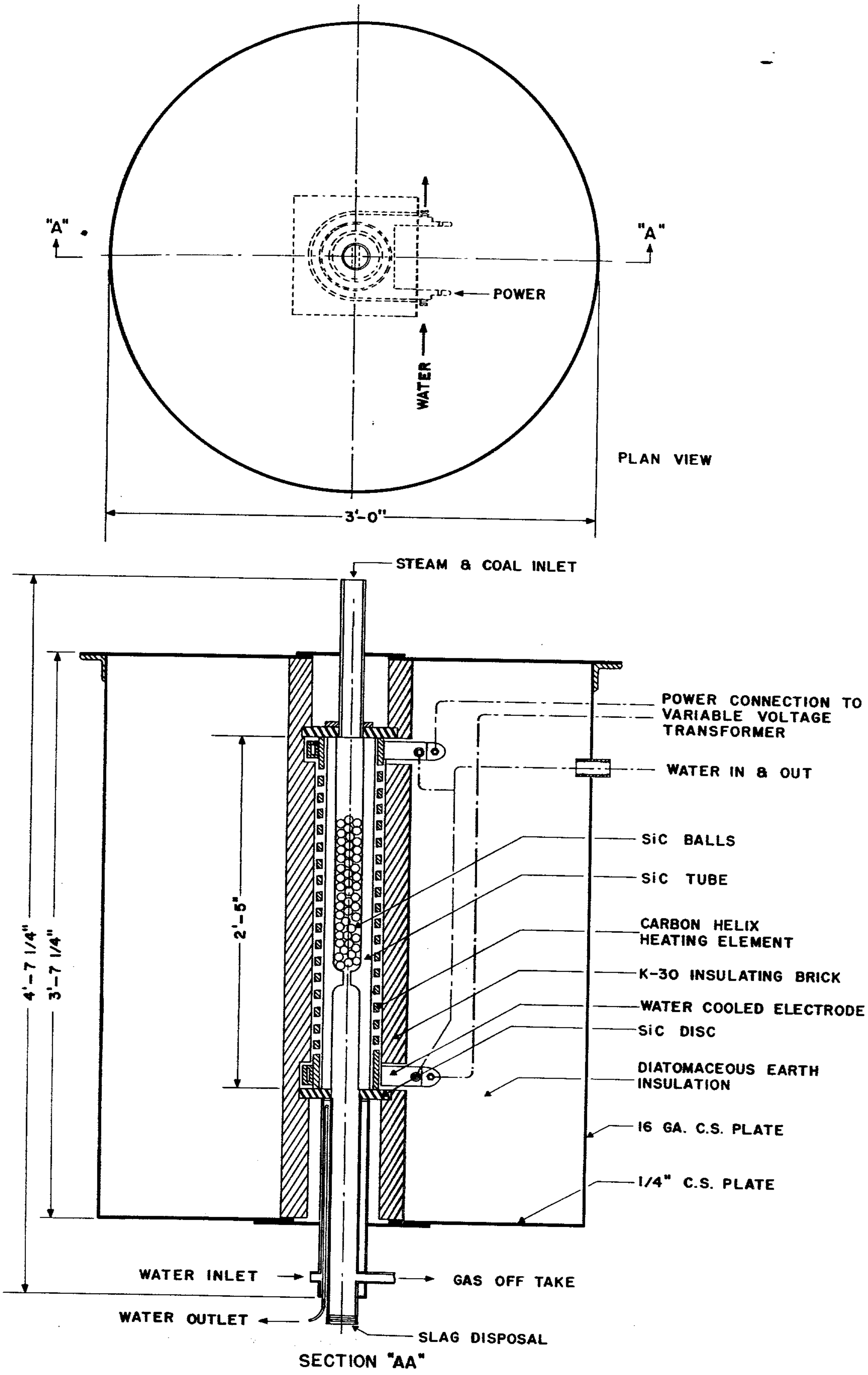


Figure 11. - Reactor for bench-scale test of nuclear gasifier materials.

should yield information on reaction rates, on reactant ratios, and on the behavior of coals of different rank and grade.

Engineering Studies and Cost Estimates

Cost estimates were made on the following phases of the program:

1. A waste-heat boiler designed for cooling the gas made in a 50-ton per hour pressure gasifier was compared with a direct-contact spray system. The estimate showed that generation of steam by waste heat was economical and that the payout time for the difference in investment between the 2 systems was less than 4 years.

2. A proposed powdered-coal air-blown gasifier for generating reducing gas from coal and air was compared with a conventional fixed-bed producer. The estimate indicated substantial saving in gas cost, provided a pulverized-coal producer is operable.

3. Cost estimates were made on three types of synthesis-gas generators, with nuclear fuel supplying the heat for reaction of the coal-steam mixture. The estimates point to a large potential saving in the cost of synthesis gas if proper design and materials of construction can be developed for operating nuclear equipment at 2,500° to 3,000° F. The estimate also suggested that the most promising design would be one in which the reactants directly contact the fuel elements.

4. At present the program of research involves pressure gasification with oxygen and also study of processes that do not require oxygen. For economic comparison of these two types of processes, the cost of bulk oxygen must be known. In cooperation with a manufacturer of oxygen plants and an operator of a large, low-purity oxygen plant, an estimate was made of the cost of producing oxygen in 200-, 400-, and 600-ton-per-day plants.

Gasification With Hydrogen

To produce a substitute for natural gas, usually referred to as high-B.t.u. gas, direct hydrogenation of coal at moderate pressures may be more economical than synthesis from carbon monoxide and hydrogen. A major advantage of direct hydrogenation would be that only about 2 volumes of hydrogen are needed per volume of high-B.t.u. gas, whereas methanation of carbon monoxide requires 4 volumes. Laboratory apparatus was designed for possible later batch experimentation, to determine reaction rates at various pressures and temperatures.

Kinetic Study of Steam-Carbon Reaction

Effects of Particle Size

Much time may be saved in developing and designing commercial-scale powdered-coal gasifiers if the mechanisms and rates of reaction between steam and carbons of different type and size are known for various gasification temperatures and pressures. Studies of this kind are being made in small-scale equipment.

The original gasification kinetics reactor was modified further to make it more nearly isothermal. Batches of sized electrode graphite were reacted with steam at 2,400° F. As in previous work, each batch was passed through the reactor tube 4 or 5 times; that is the weighed residue from the first pass was recycled for the second pass, and so on.

Particle-size ranges investigated were -60- to +80-, -80- to +100-, -100- to +120-, -120- to +140-, -140- to +170-, -170- to +200, and -200- to +230-mesh, U. S. Sieve Series. Particles larger than 60-mesh could not be used owing to their short residence time, while the particles smaller than 230-mesh fell in a turbulent pattern and not in streamline flow, as desired.

In all runs a higher rate of reaction was found for the first pass than for succeeding passes (fig. 12). The rate of reaction of graphite with steam appears to be a function of the particle size, the rate increasing as the particle diameter decreases.

High-Temperature Kinetics Reactor

Reactions in large-scale gasifiers take place considerably above 2,500° F., the highest temperature investigated in previous kinetic studies, since temperature of fuel particles in the oxygen-fed flame may rise well above 3,500° F. The rate of gasification in the temperature range 2,500° to 3,000° F. is of interest to operators and designers of large-scale gasifiers.

A high-temperature reactor for isothermal operation at temperatures up to 3,500° F. is under construction. The reaction chamber will be a 1/4-inch-thick, 9-foot-long alumina tube of 2-1/2-inch I.D. and will be heated by a 4-inch-I.D. carbon tube used as an electrically heated resistor (fig. 13). Electric-power input will be controlled by a tungsten-molybdenum thermocouple. The controller will operate a voltage-regulating assembly connected to a transformer that will supply current to heat the carbon resistor tube. The carbon heating element, insulated by 2 feet of lampblack, will be contained in a 5- by 8-foot brick furnace provided with external, sectional heating.

Flashing Coal-Water Slurries in Heated Coils

Variables in Preparing Suspensions of Micronized Coal in Steam

The object of this study is development of a controlled method for flashing a coal-water slurry (in an externally heated alloy-steel tube coil) into a suspension of fine coal in steam. Present objectives are to establish the effects of variables on micronization of the coal and on the rate of delivery of the coal-steam suspension. The apparatus is shown in figure 14. Two runs were carried out with 100° F. difference in the temperature of the coil-outlet (fig. 15). The temperatures for runs SPSM-4 and SPSM-2 were 550° and 655° F. At 50-percent cumulative mass fraction, particle sizes were: For the feed coal, 180 microns; for products from run SPSM-4, 160 microns; and for products from run SPSM-2, 90 microns. The average particle size recovered in run SPSM-1 from the baffle chamber was 130 microns and that removed from the cyclone and bag filter was 60 microns. The weighted average particle size in run SPSM-1 was 124 microns.

Gasification of Coal-Water Slurries Without Oxygen

Efforts are underway to gasify coal by methods in which the heat required for the reaction is supplied from an external source, thereby eliminating the use of oxygen. In one such method coal-water slurries are heated to reaction temperatures within an electrically heated coil. Such a process might utilize waste heat from the graphite piles of an atomic energy installation or heat generated especially for this purpose by nucleonic fission reactions.

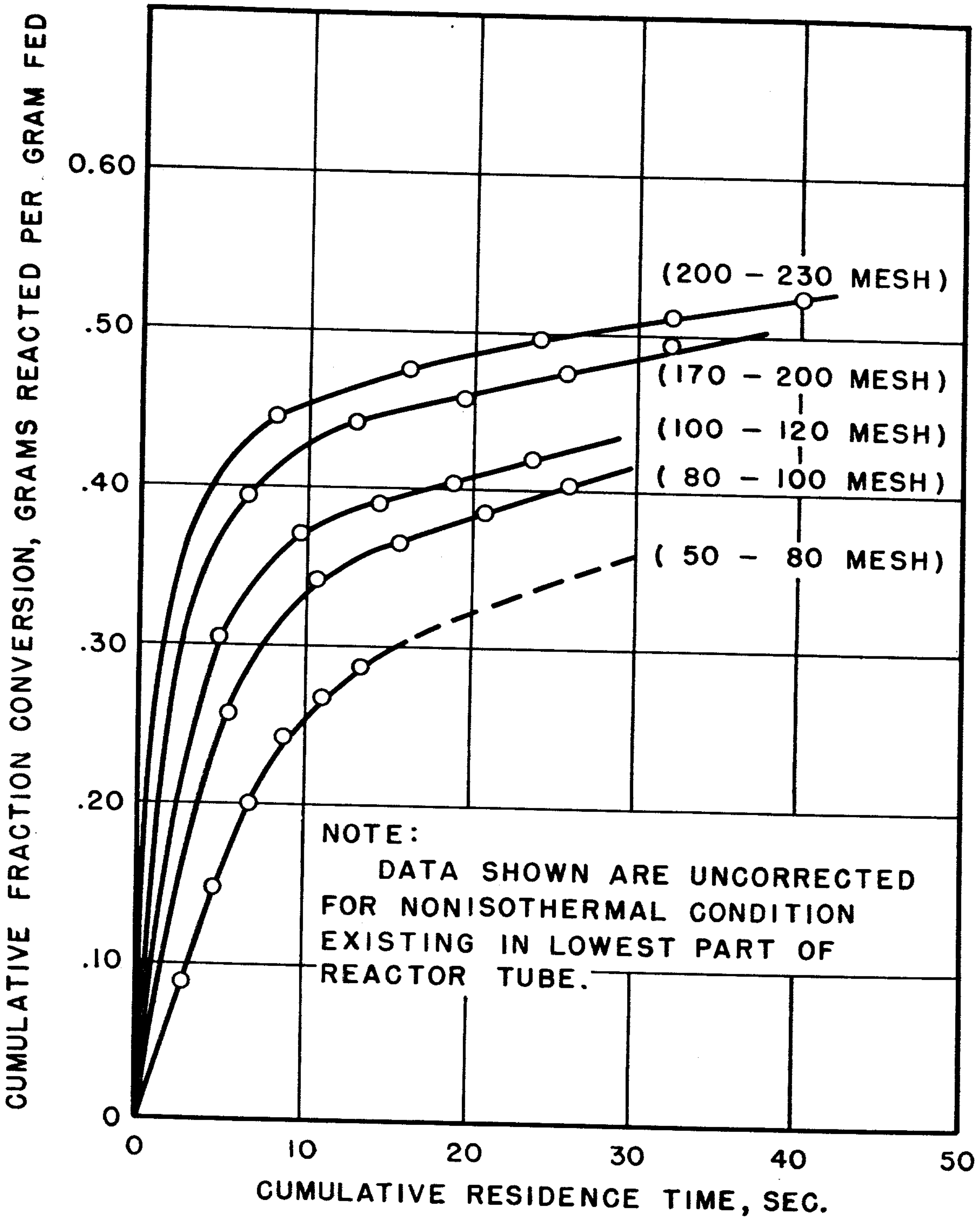


Figure 12. - Reaction of electrode graphite with steam at 2,400° F. in an isothermal reactor; effect of contact time and particle size on carbon conversion.

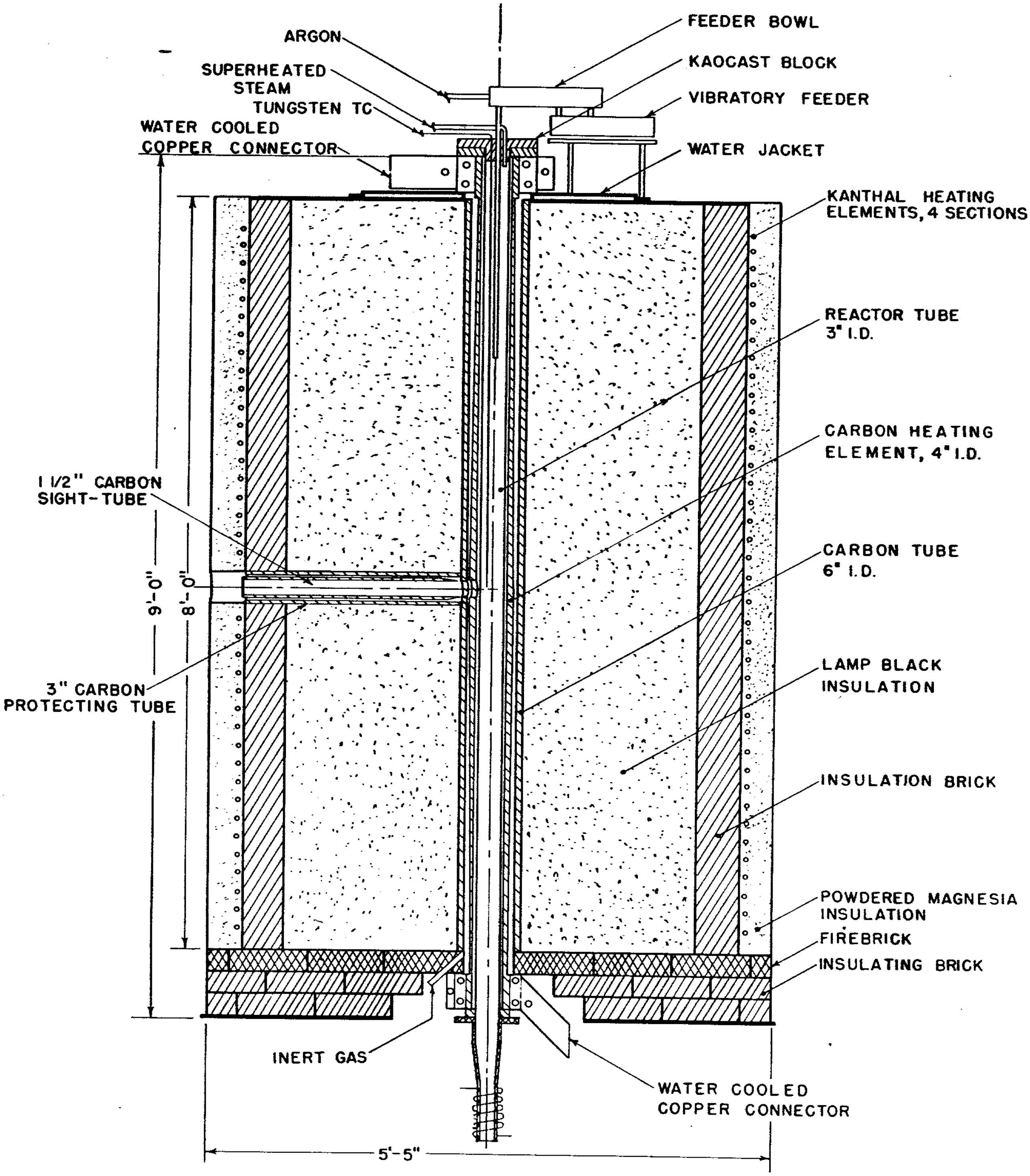


Figure 13. - Isothermal reactor for the kinetic study of carbon gasification at high temperatures.

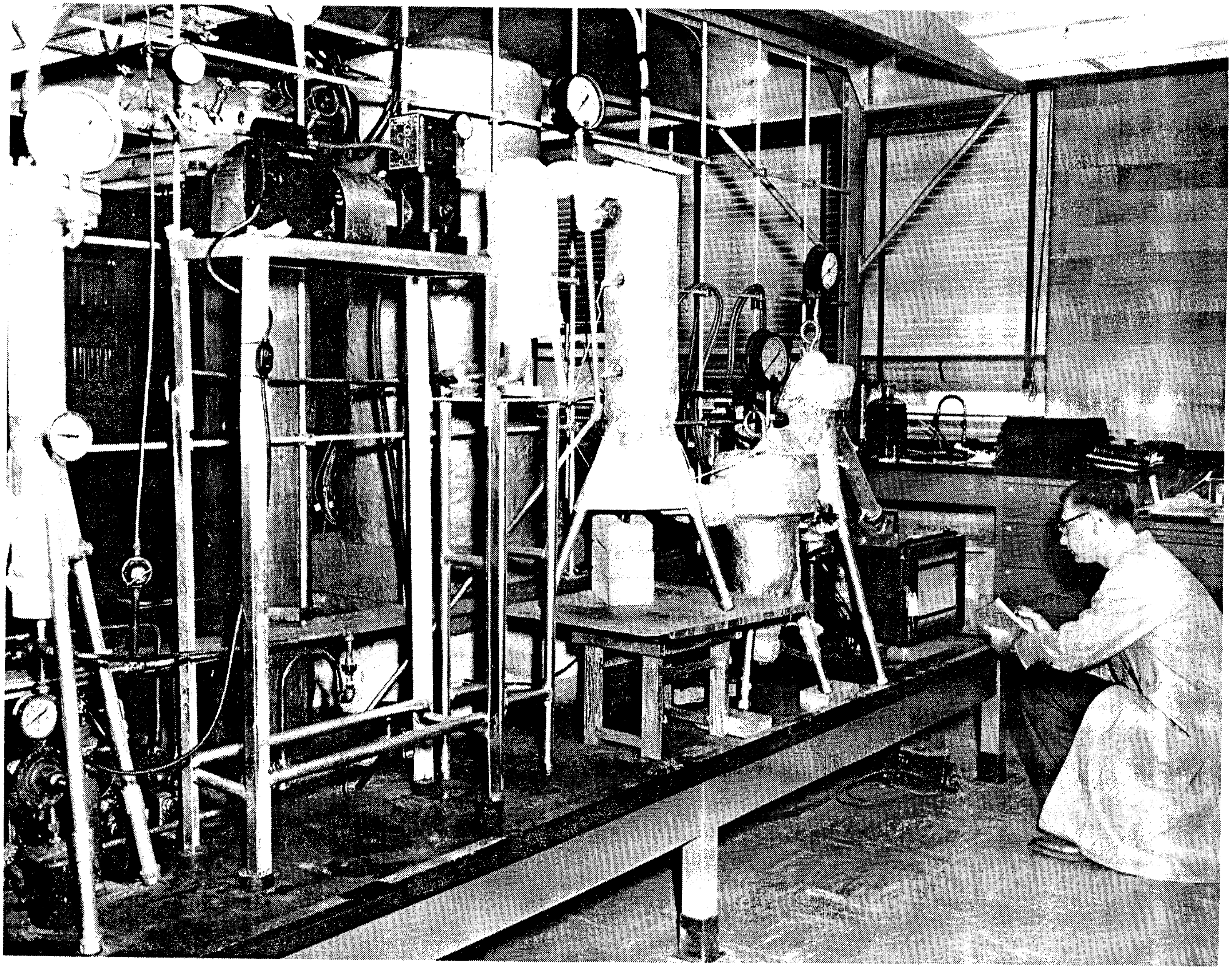


Figure 14. - Apparatus for micronization of coal by flashing coal-water slurries in heated coils.

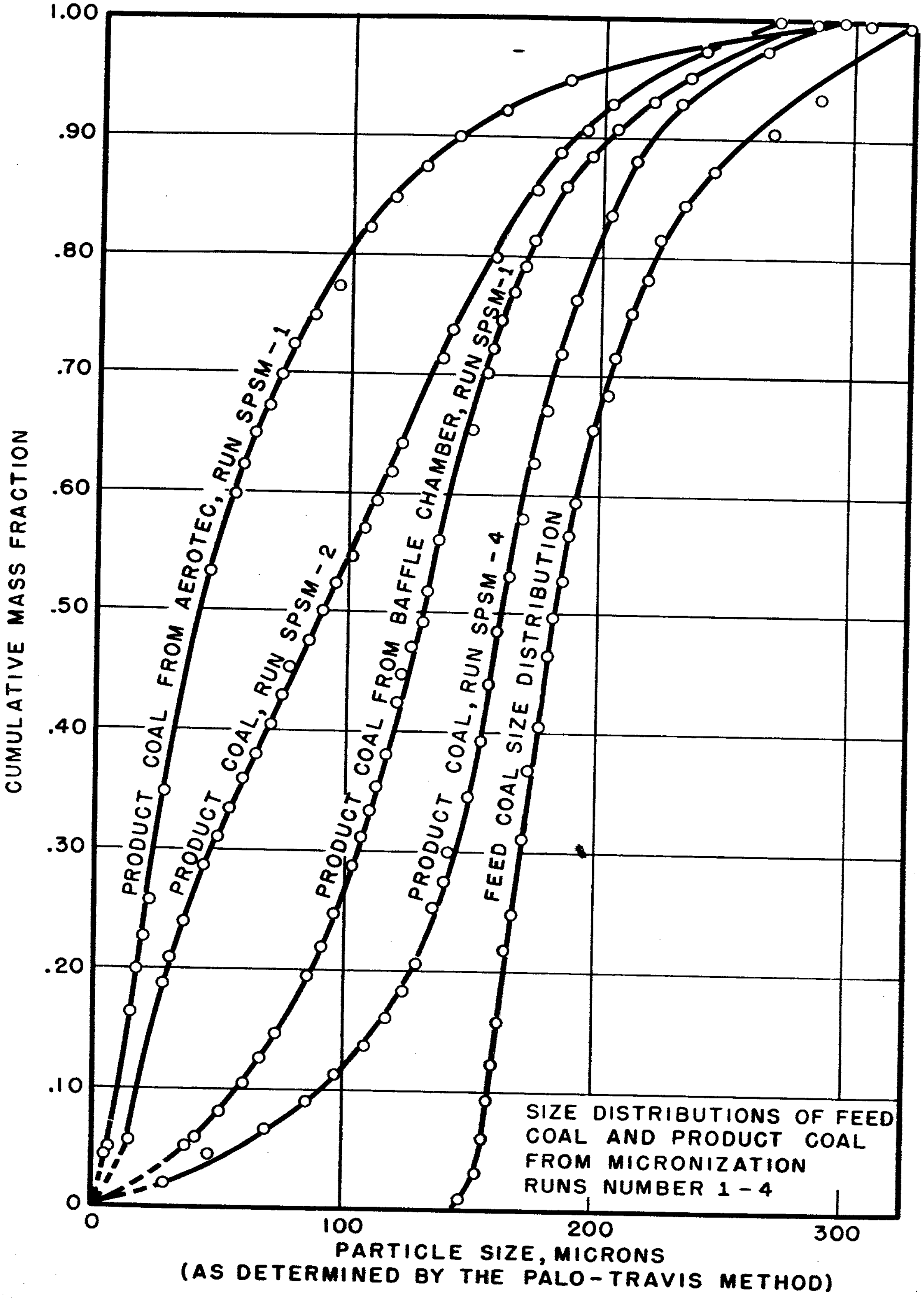


Figure 15. - Micronization of coal by flashing coal-water slurries in heated coils.

A coiled-tube reactor and accessories for flash-gasification of coal-water slurries are shown in figures 16 and 17. The coal-water slurry is pumped at constant rate into the 117-foot-long, 3/8-inch, 25Cr-20Ni alloy-steel tubing of 9-inch coil diameter. The coil, placed within an insulated and gastight brick furnace maintained at 1,500° F., is electrically heated, acting as the resistor. Argon is passed slowly through the insulation of the furnace to protect the coil from exterior oxidation.

Six preliminary runs have been made at 1,350° to 1,850° F. At 1,850° F. outlet-gas temperature, approximately 65 percent of the carbon in Lake deSmet, Wyo., subbituminous coal was gasified, with a contact time of only one-tenth second. The gas contained about 16 percent carbon monoxide, 60 percent hydrogen, 20 percent carbon dioxide, and 3.5 percent methane. Less gasification was obtained at lower temperatures: 14.7-percent carbon conversion at 1,350° F. and 18.5 percent at 1,550° F. outlet-gas temperatures.

Sorption and Recovery of Oxygen From Air

The object of this work is to develop an economical process for separating oxygen from air by its selective sorption on a chemical oxygen-carrier and subsequent recovery by desorption.

A bench-scale glass apparatus was constructed for preliminary screening of potential oxygen carriers. An improved model now in use, provided with a diaphragm-type gas pump for cycling oxygen or air during the sorption step, is shown in figure 18. Polarographic studies have been made also to determine the ease of sorption of oxygen by salcomines, Versenes, and other types of metal chelates. The data confirmed the results of direct sorption tests in the glass apparatus. None of the Versene-type chelates had oxygen-carrying properties comparable with those of salcomine (cobalt disalycylal ethylenediimine).

To compete with the currently used rectification of liquefied air for production of oxygen, a new sorption process should (1) require a relatively small temperature difference between absorption and desorption steps; (2) have low heats of reaction for the oxygenation and reduction steps; (3) absorb oxygen from the air and desorb it from the compound at pressures not far from atmospheric; (4) involve simple apparatus so that design and construction of a commercial plant may be economical; (5) cause virtually no deterioration of the compound itself on cycling. Of the compounds thus far tested, the salcomine-type chelate meets all of the conditions listed above except the last one, that is minimal deterioration on cycling. Salcomine deteriorates very little, yet sufficiently to make the process economically noncompetitive. The immediate problem is to prevent or considerably reduce this deterioration or to find another suitable compound with no appreciable deterioration.

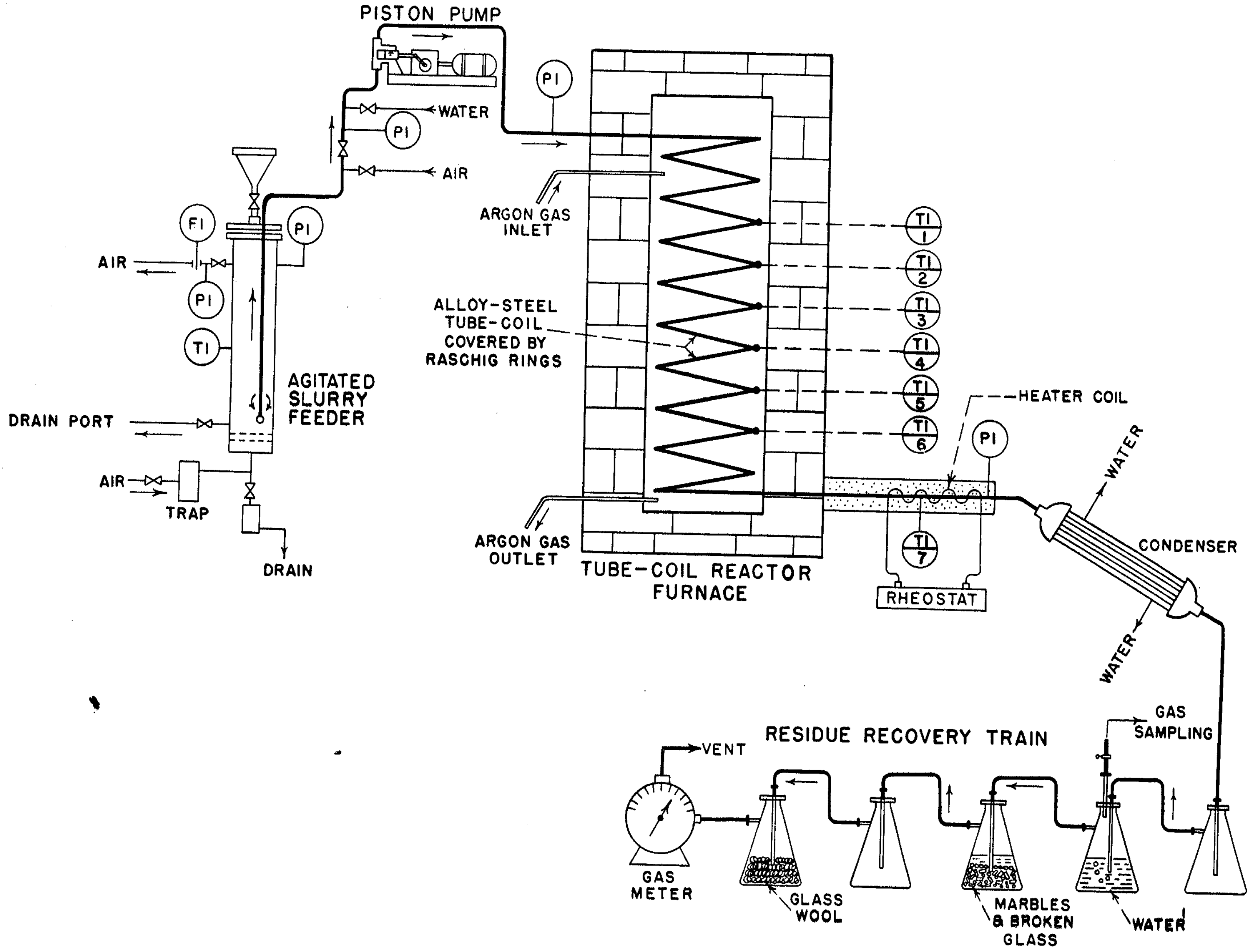


Figure 16. - Flow diagram of tube-coil reactor furnace and accessories for gasifying coal-water slurries.

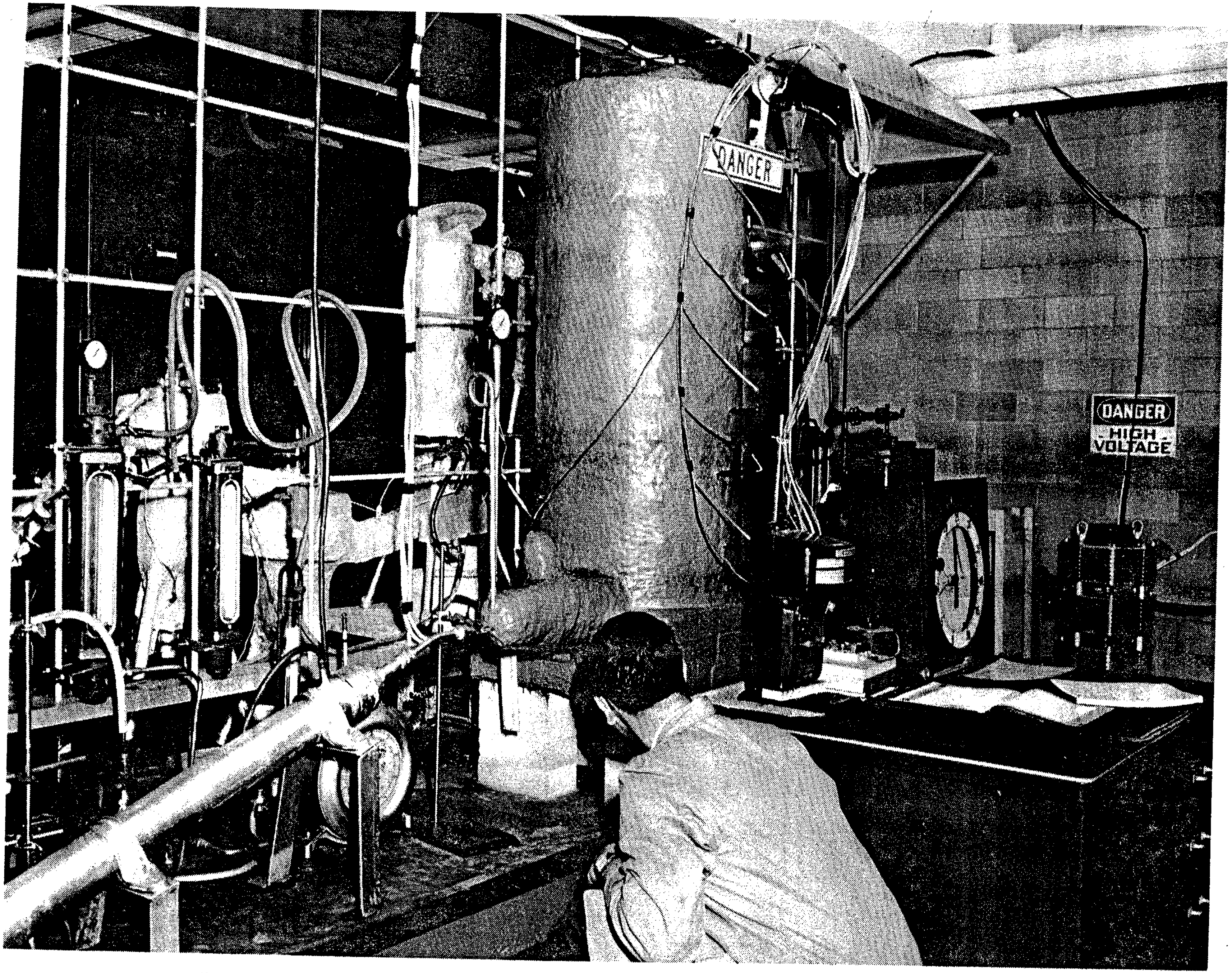


Figure 17. - Tube-coil furnace and accessories for gasifying coal-water slurries.

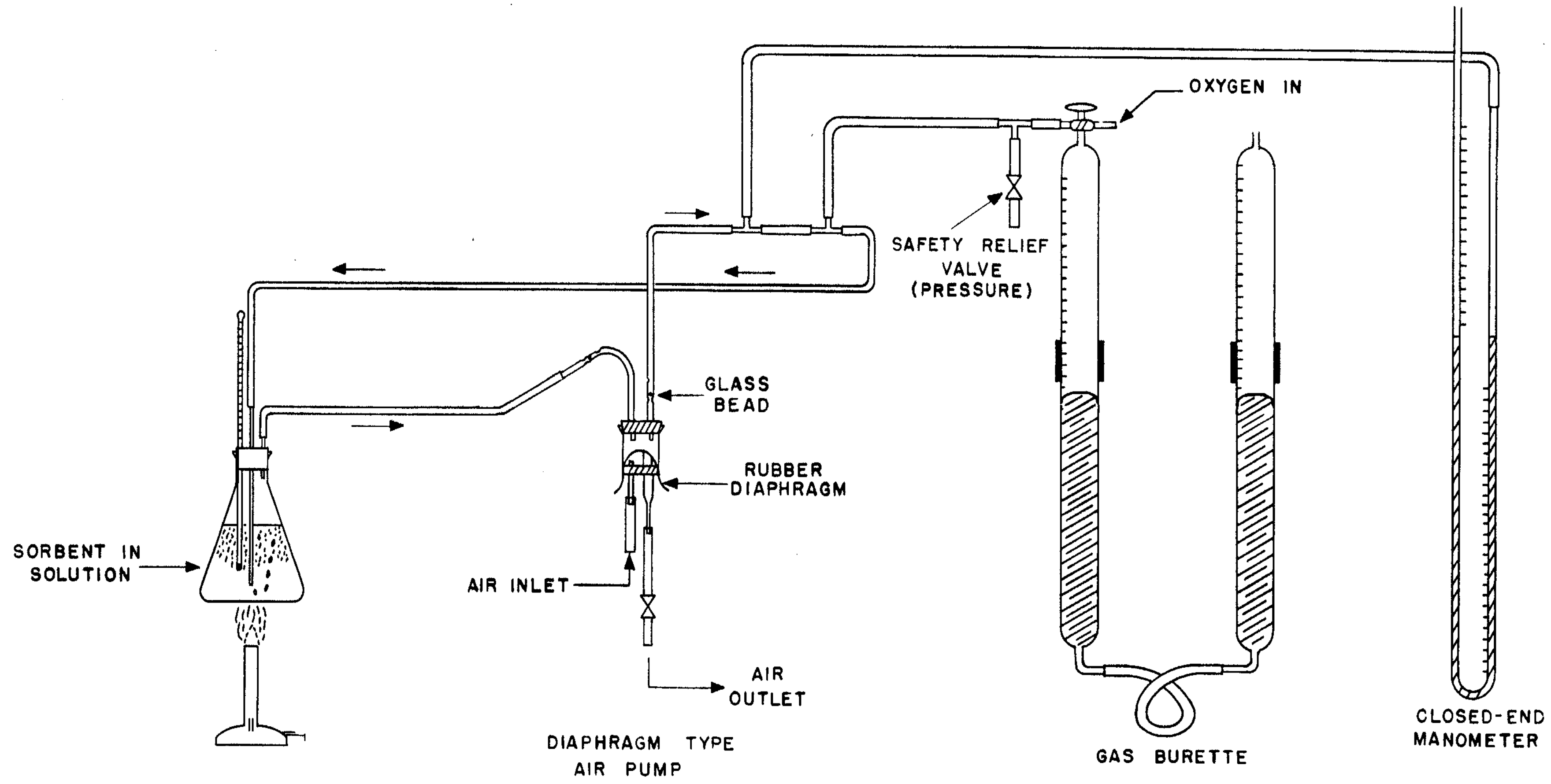


Figure 18. - Improved apparatus for preliminary oxygen sorption-desorption tests.