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GAS RECOMBINATION SYSTEM OF THE LOS ALAMOS HOMOGENEOUS REACTOR

by

M. E. Bunker[1]
R. P. Hammond[2]
L. D. P. King[1]
J. A. Leary[2]
W. R. Wykoff[2]

[1] Members of Group P-2
[2] Members of Group CMR-10

PHYSICS

I. Introduction

The Los Alamos Water Boiler is the first reactor in which fission fragments are released in the moderator itself. Other reactors using liquid moderators have had only ionizing radiations in the form of neutrons, beta rays and gamma rays in the solution. In the case of the water boiler the large energy release in the fission process causes considerable additional decomposition of the moderator as well as a great increase in the radioactivity of the released gases.

Operation of the early Hypo model [1] of the reactor at a specific power of 0.4 kw/liter of solution made it evident that it would be advantageous to recombine as many of the dissociated products as possible. This would reduce the changes in solution composition during operation and simplify the disposal of the remaining radioactive gases. When the reactor was modified for higher power operation, provisions were made for the installation of a gas control system. Initial operation of the present Supo model [2] at a specific power of 2.4 kw/liter of solution indicated the advisability of installing a recombination system as soon as possible. The solution decomposition was as high as 4.5 cc/min at 80°C. This represents a hydrogen release of almost 8 liters*/min. In addition to this potential explosion hazard, frequent solution additions were required to maintain satisfactory reactor performance. The large volume of highly radioactive gas also made it difficult to maintain satisfactory backgrounds at all times on counting equipment in the neighboring technical areas.

It is the purpose of this report to give a description of the equipment and initial tests of a recombination system which has been installed on the reactor. This equipment has given excellent results in the first eight months of operation.

II General Design and Layout

A. Design Criteria. Two methods were available for effecting the recombination of hydrogen and oxygen in the reactor off-gas: flame combustion and contact catalysis. A flame combiner would have the advantages of low pressure drop and unlimited lifetime, but since the water boiler is operated at widely varying power levels with frequent shutdowns, it appeared that steady operation of a flame combiner would be difficult to attain. For the catalytic method the principal difficulties to be anticipated were plugging and poisoning.

Platinized catalyst materials of high activity for the hydrogen-oxygen reaction were commercially available on either charcoal or ceramic support. Previous experience indicated that repeated heating and cooling of charcoal material tended to produce fines which

[1] Rev. Sci. Inst. 22, 492 (1951).

[2] LA-1301.

* Gas volume measured at an altitude of 7000 ft, 23 in. Hg barometer, 25°C.

would plug the bed. The ceramic support was therefore indicated as the preferable choice.

The water-boiler off-gas stream is not pure hydrogen and oxygen, but carries gaseous fission products, water vapor and small amounts of entrained reactor solution containing uranyl nitrate, fission products, and free nitric acid. The possibility of catalyst poisoning and bed plugging from these materials had to be considered.

Laboratory tests of platinized alumina pellets in electrolytic gas streams containing water and nitric acid vapor showed high catalytic activity except for a temporary loss when the pellets were actually wetted by condensed water. The effects of radiation and fission products could not be studied within the allowable time, so it was decided to proceed with a design which would at least minimize such effects. Design criteria were set as follows:

1. A rapidly circulating gas system to keep both the reactor and stack line below the explosive concentration of hydrogen.
2. Removal of as much entrained liquid as possible from the gas stream, and return of this matter to the reactor solution.
3. Cooling of the gas leaving the reactor to minimize the nitric acid evaporation.
4. Use of a large quantity of catalyst to minimize poisoning effects.
5. Use of two separately valved and separately removable catalyst chambers, so that the second could be exposed to the gas only upon the exhaustion of the first.
6. Instrumentation to detect any change in flow or resistance of the system and to indicate the active zone of the catalyst bed itself. A progressive movement of this hot zone would indicate a progressive poisoning of the catalyst.
7. A catalyst chamber designed with an internal heater to preheat the catalyst and drive out condensation on start-up, if necessary.
8. Condensation and return of recombined water to the reactor, with provision for diverting known quantities of this water to waste when desired.
9. A circulating blower with a gas-sealed shaft and an external, readily serviced drive. When supplying a sweep gas rate of 100 liters/min, the blower should have a pressure rise of 8 in. of water.
10. The entire system to operate slightly below atmospheric pressure.
11. The entire system to be constructed of type 347 stainless steel.

B. Schematic Layout. A block diagram of the circulating gas system is shown in Fig. 1. The off-gas, containing primarily hydrogen, oxygen, nitrogen, gaseous fission products, water vapor, nitric acid vapor and entrained material, is cooled in the long reflux condenser. Both the condensed vapors and de-entrained liquid drain back into the reactor. An after-

filter is provided to remove any residual entrainment and to reduce to a minimum the foreign material carried to the catalyst. The blower provides sufficient pressure to circulate 100 liters/min of gas, the actual flow being indicated by an orifice near the blower inlet. The manifold, following the blower, leads the gas to either of the two catalyst chambers. This manifold, being the point of highest static pressure in the system, is also vented to an exhaust line and stack. The stack pressure becomes the reference pressure of the system; all other parts of the system assume pressures negative to the stack. The maximum pressure in the system is held at 3 in. of water below atmospheric by a blower in the exhaust stack. This partial vacuum is advantageous in preventing gas leaks into the reactor room.

In passing through the catalyst the gas is heated by the recombination reaction. After leaving the catalyst the gas is cooled to condense the converted water and reduce the heat load on the reactor cooling system. A calibrated trap in the return line fills with water and can be dumped to waste when desired.

C. Components. A more detailed description of the various components of the recombiner system is given in the succeeding paragraphs. The only metal used in fabricating the entire system was type 347 stainless steel. It was known from earlier experience that this metal was highly resistant to corrosion by nitric acid, and that welds in the metal could be expected to withstand severe corrosive action indefinitely. The intercomponent piping is made of 11/16 in. I.D. stainless steel tubing, with either 1/32 or 1/16 in. wall thickness.

1. Reflux Condenser Unit. The chief design consideration in regard to the reflux condenser unit was that it must cool the sphere exit gas down from about 75°C to below room temperature. Furthermore, the condenser unit had to be made so that it would fit into the existing stack mounted on the sphere. The first trial model of the coil, made of copper, consisted of a double helix, the two helices being of the same diameter and joined at the lower end. Thus the cooling water traveled down one helix and up the other. Although this model produced the desired temperature drop, it was discarded in favor of a single helix (described below) for the following reasons: (1) A single helix was found to give even better temperature characteristics, due to the fact that the gas near the exit was not in contact with the exit water line, (2) a single helix was much easier to form. The pitch of the helix was made so that the condensate would run freely down the coil and not hang in stagnant drops. This feature was checked visually in a mock setup by simulating the outer wall of the condenser with a section of glass tubing.

The reflux condenser unit is shown in detail in Fig. 2. The gas from the after condenser enters the unit at A and passes down the central 3/4 in. tube into the sphere. The disk at the bottom of this tube deflects the incoming gas in such a way that it sweeps slightly above the surface of the solution fairly uniformly and with minimum surface disturbance. The circulating gas plus the reactor off-gases then pass upward over the cooling coil, which serves to condense the water vapor and acid fumes contained in the gas. The condensate, of course, drips back into the sphere. The cooled gases leave the unit at B and pass on to the entrainment trap. The cooling water enters at C and leaves at D, flowing counter-current to the gas flow for more efficient cooling. The cooling coil and return line are made from a single continuous 15 ft length of 1/4 in. O.D. by 1/32 in. wall stainless steel tubing. The 1/4 in. tube extending out the bottom of the 3/4 in. central tube is a bubbler line used to define an upper limit to the solution level. The portion of the outer section of the condenser above the exit opening is filled with a solid piece of stainless steel which acts both as a radiation shield and as a block to keep the radioactive gas from rising any higher than necessary.

2. Entrainment Trap. An entrainment trap is located ahead of the blower to stop any entrained liquid carried over from the reflux condenser. This unit can be seen clearly in Fig. 10. It consists of a 3-1/2 x 3-1/2 x 17 in. rectangular stainless steel box filled with about 125 gm of fine stainless steel wool. The box is tilted so that any trapped liquid will run back into the sphere. There was some worry that there would be a prohibitively high pressure drop in the trap when and if the steel wool got saturated with water. However, when measurements were made at a gas flow of 100 liters*/min, the trap was found to have a pressure drop of only 0.25 in. of water after steam had been passed through the trap for 10 min.
3. Circulating Blower. Restrictions imposed by the reactor stacking and the requirement that all parts of the system exposed to the circulating gas be stainless steel necessitated the design and fabrication of a special blower for the system. The design criteria for the fan were that a pressure head of 8 in. of water be maintained while flowing 100 liters/min of gas and that the unit be tight against gas leakage.

* Gas volume measured at an altitude of 7000 ft, 23 in. Hg barometer, 25°C.

The blower used at present is a conventional design of the high-speed centrifugal type, the unique feature being the graphite shaft seal used to prevent leakage of radioactive gases. The impeller is 9 in. O.D. and has 8 straight blades 3/4 in. wide. A graphite-to-graphite shaft seal backed up by controlled air pressure prevents the leakage of contaminated gas from the system. A new design of the driving mechanism is under construction which uses graphite bearings in place of ball bearings and eliminates this air-pressure seal by the use of a sealed-rotor motor. The new design also allows much easier access to the blower bearings. Figure 3 is a sectional view of the entire blower assembly showing the newly designed blower in its shield can. Tests were conducted to measure the pressure rise in the blower as a function of fan speed. Performance was obtained with two potential sweep gases, helium and air. The results for air are shown in Fig. 4. At a speed of 4720 rpm the fan produced a pressure rise of 8.2 in. of water when pumping 100 liters/min of air. At this flow rate the pressure rise produced when circulating helium was 1/5 that obtained with air.

4. Catalyst Chamber. Laboratory tests on a proposed catalyst chamber revealed the following:

    a. Internal bed heaters were unnecessary.

    b. Pressure drop data taken with the catalyst bed at room temperature were invalid. When hydrogen and oxygen were being combined in the catalyst bed, the increase in temperature caused an increase in gas viscosity and velocity. This resulted in a serious increase in pressure drop across the catalyst chamber. In order to minimize this effect the catalyst bed cross section to gas flow was maximized to improve heat distribution as well as to reduce gas velocity. The improvement in heat distribution caused a greater heat loss to surroundings, thereby reducing the fractional increase in pressure drop at high temperature, while the reduction in gas velocity resulted in a lower initial pressure drop.

The final chamber design was simply a rectangular can of 6-1/8 x 6-7/8 x 5 in. inside dimensions, as shown in Figs. 5 and 5A. Gas flows through the central tube to the plenum in the bottom of the can and is thereby preheated after initial start-up by the heat of the reaction in the catalyst bed.

It is then distributed through a perforated plate and passes up through 1700 gm of catalyst material supported on 18-mesh stainless steel screen. The gas leaving the catalyst bed is filtered through stainless steel wool in the exit plenum to prevent any possible catalyst dust from reaching the reactor. The catalyst material was purchased from the J. Bishop and Co. Platinum Works, Malvern, Pa., and consisted of 1/8 in. dia. x 1/8 in. long alumina pellets coated with platinum. The pellets are 0.3 per cent by weight platinum. Three thermocouple wells were installed in the catalyst bed as indicated in Fig. 5A by thermocouples T_2, T_3 and T_4. With a new catalyst bed the maximum temperature is obtained at the gas inlet side of the bed (T_2). However, if the catalyst in this area deteriorates, the primary reaction zone will move deeper into the bed. It was anticipated that when the temperature indicated by T_3 became less than that of T_4, the catalyst bed would be considered nearly spent. The inlet gas temperature (T_1), the outlet gas temperature (T_5) and the temperature of the catalyst shield pot (T_6) were also recorded. A 550-watt Chromolox heater was installed under the catalyst chamber for drying the catalyst bed should the need ever arise.

The chamber was tested in the following manner: For each test known rates of hydrogen, oxygen and sweep gas were passed continuously through individual drying and metering stations, then through the catalyst chamber. The exit gas from the chamber was then cooled to $10^{o}C$ and the total rate of water formation (condensate + humidity) was determined. Comparison of this with the known rate of hydrogen introduction gave the recombination efficiency for a single pass. Recombination efficiency was also determined by hydrogen analysis of gas samples taken from the catalyst chamber inlet and outlet. Temperature-vs-time plots were continuously recorded by means of thermocouples installed in the catalyst bed and the entering and leaving gas lines. Pressure drop vs time was also recorded. A typical temperature-vs-time plot is shown in Fig. 6. Apparently, thermal equilibrium was attained 1 hour after the start of hydrogen introduction. Since the inlet side of the catalyst bed attained the highest temperature, most of the reaction occured in this zone. A plot of pressure drop vs exit gas temperature across the catalyst chamber is shown in Fig. 7. The increase in pressure drop with gas temperature is due to increases in both gas velocity and gas viscosity. Good data for the recombination efficiency were not obtained;

values ranging from 80 to 103 per cent recombination for a single pass were obtained by both methods. Due to scheduling of reactor operation, there was not sufficient time to obtain better data.

5. After Condenser. The design requirements for the after condenser were that it should be able to reduce the temperature of gas flowing through it at the rate of 100 liters/min from about 350°C to at least the sphere temperature of 70°C, and at the same time to condense water at the rate of 4 cc/min of condensate. By experimenting with brass models, it was soon found that a very small condenser (2-1/8 in. diameter x 11 in. long) would be adequate, and that a cooling-water flow of as little as 0.2 gal/min was sufficient to bring the exit-air temperature down to within 15°C of the inlet water temperature. Furthermore, it was found that a double helix cooling coil was much more efficient than a single helix, presumably due both to the increased coil area and the increased turbulence induced in the gas.

   The condenser is shown in Fig. 8. Its construction is very simple. The hot gas from the catalyst chamber enters at A, comes in contact with the cooling coils and leaves at a reduced temperature at B. The condenser was installed with a slight tilt toward the exit end so that the condensed water would run into the sphere. The centrally located baffle was put in to increase the turbulence of the gas. Similarly, the tube along the axis with the closed-off ends was included merely to take up dead space and thus prevent streamline flow in that region. The two cooling coils are made of 1/4 in. O.D. stainless steel tubing. They have a common inlet and outlet, the water flowing in a direction opposite to the gas flow for most efficient cooling. The sleeves around the water lines where they enter and leave the condenser were used because (1) it was felt there would be less danger of burning holes in the water lines during welding, and (2) with the sleeve arrangement such holes would be completely outside the condenser and hence would be easier to find and to fix.

6. Liquid Trap. A liquid trap is located between the after condenser and the reflux condenser. This trap consists merely of a 12 in. section of 3/4 in. tubing with a valve at the bottom which opens into a dump line. It is shown clearly in Fig. 10. The trap holds about 82 cc and is normally full of water when the recombiner system is operating, since this amount of

water is recombined in the catalyst chamber in a matter of about 20 min if the reactor is at a power of 25 kw.

7. Overpressure Relief Valve. In parallel with the gas bleed valve to the exhaust line is a solenoid valve which opens automatically if the pressure in the recombiner system exceeds atmospheric pressure. This is merely a safety feature to eliminate pressure build-up in the system and to reduce the possibility of leakage of radioactive gas. The pressure indicator which actuates the solenoid is also connected to one of the reactor safety circuits so that the pile is automatically shut down if the system pressure exceeds atmospheric pressure.

III. Preliminary Testing of Assembly

It was felt desirable to make a complete mock-up of the recombiner system, using the final components where feasible, and simulating expected operating conditions as nearly as possible. Therefore, after all the components had been fabricated, the system was assembled on a bench outside the reactor. A view of this test setup is shown in Fig. 9. The stainless steel sphere and adjoining stack were full-size models of those in the reactor. The sphere was filled with a nitric acid solution which had the approximate pH of the actual boiler solution. The acid solution was kept at a temperature of 80°C during most of the tests by means of a heating mantle placed under the sphere. The numerous lines of small tubing appearing in the photograph are pressure lines which go to Magnehelic [3] pressure gauges. The white covering over some parts of the system is asbestos insulation.

Since separate tests had been made on the operation of the catalyst chamber, it was decided that use of hydrogen in the mock-up was unnecessary. This eliminated the difficulty of devising a safe way to generate the required amount of hydrogen within the system. Then, to simulate the recombination process, the catalyst chamber was replaced by a combination heater and steam injection unit. This unit was made so that it had the same pressure drop as the catalyst chamber at a gas flow of 100 liters/min.

When the mock-up system was operated, it was found that all components behaved satisfactorily and that no major changes were needed. The pressure drop in the system was found to be higher than anticipated, due mostly to the fact that the pressure drop across the reflux condenser was 25 per cent higher when the coil was wet than when it was dry. It developed that a time of 20 to 30 min was required to attain pressure equilibrium in the system. At pressure equilibrium, a blower speed of 4720 rpm was found necessary to maintain a flow of 100 liters/min.

[3] Made by F. W. Dwyer Mfg. Co., Chicago, Ill.

IV. Reactor Installation and Performance

A. Component Arrangement. The installation of the recombiner components into the reactor was fairly straightforward since during the modifications from the Hypo to the Supo version [2] consideration was given to the possible future installation of a gas disposal system. The reflux condenser replaces a removable level indicator unit and the catalyst chambers and blower are placed in shielded boxes which replace concrete shield blocks. Figure 10 is a phantom view of the complete installation. Figure 11 is a photograph of the system taken during assembly.

Pressure drops and gas flow rates are transmitted for observation and recording at the control panels by means of condenser pressure gauges[4]. Magnehelic gauges are used to give direct pressure readings at the reactor. Temperatures at various points in the system are measured by means of thermocouples. These are recorded on a 0-600°C or a 0-100°C Brown Recorder. A drinking fountain type water cooler supplies the cooling water for the two condensers, which are connected in series. A by-pass valve permits independent water adjustments on the after condenser.

B. Operational Behavior. For the initial operation the reactor power was gradually increased while a close check was maintained on the catalyst bed temperatures. The first indication of gas recombination appeared at a power of 300 watts when a slight rise of temperature was observed on the bottom thermocouple (T_2) (Fig. 5). When the power was increased to 1 kw all portions of the catalyst bed rose at a rate of about 1°C/min. Any further increase or decrease in reactor power appeared on the Brown Recorder with only a slight time delay. This quick response at room temperature indicated that one probably would not have to use the heater shown in Fig. 5A. From these initial tests through the present eight months of operation the entire system has performed very satisfactorily. No detectable change has occurred in the catalyst bed and the heaters have never been used.

In order to keep all gas contamination from the pressure indicators and the gas sampling valves, an air bleed-in system was installed whereby a few cubic centimeters of air per minute can be bled into each of the tubes. It takes about 24 hours for activity to appear at the tube ends with the bleed-air shut off, and less than 1 cc/min is sufficient to keep all radioactivity out of these lines.

Typical pressure drops and temperatures in the recombination system for a 25 kw reactor power level are as follows:

Gas circulation rate	.41" water orifice pressure drop or 100 liters/min
Catalyst chamber drop	2.2" water

[4] These gauges are designed to resist corrosion and pressure surges and will be described in a more detailed report on the reactor than is given in Reference 2.

Reflux condenser drop	4.5" water
Maximum system pressure	-3.1" water
Exhaust line pressure	-3.2"
Catalyst bed temperature, inlet region	467°C
Catalyst bed temperature, middle region	438°C
Catalyst bed temperature, outlet region	369°C
Reactor solution temperature	75°C
Reflux condenser water flow	.17 gal/min
Reflux condenser water temperature in	3.5°C
Reflux condenser water temperature out	20°C
After condenser water in	20°C
After condenser water out	28°C
Air temperature out reflux condenser	15°C
Air temperature into catalyst chamber	45°C
Air temperature out catalyst chamber	211°C
Air temperature into after condenser	211°C
Air temperature into sphere	33°C

Two types of catalyst chamber behavior have been observed when the inlet bed approaches a temperature of 580°C. At this temperature the hydrogen and oxygen mixture appears to ignite. If the reactor is run at powers in excess of 38 kw so that the rate of temperature rise in the catalyst bed is fairly rapid on approaching the above temperature, the inlet bed shows a sharp drop in temperature while the outlet air temperature shows a more rapid rise. The other bed temperatures and the catalyst pot itself decrease in temperature. These effects are probably due to the ignition of the hydrogen and oxygen in the catalyst region above the supporting screen. If the reactor is run at 35 to 36 kw so that the rate of rise of the catalyst temperature is very gradual when 580°C is reached, a slightly different phenomenon is observed. The inlet bed temperature decreases, but less rapidly than in the case above, while the air outlet temperature shows a decrease. The catalyst pot temperature, on the other hand, shows a more rapid increase. This may be due to a burning of the gas in some region preceding the catalyst pellet region. When the reactor power is reduced to 25 kw, the catalyst chamber reverts to normal operation.

C. Determination of Hydrogen Formed Radiolytically. It was of interest to determine the hydrogen evolution of the reactor as soon as possible. Actual chemical analyses of the gas obtained from various points in the system were planned, but such experiments required considerable preparation. Since previous testing had indicated that the catalyst

chamber temperatures were very dependent on the hydrogen concentration for a given rate of sweep gas flow, the following experiment was done to determine the rate of radiolytic hydrogen formation:

1. With the recombiner system at normal operating conditions and the reactor at zero power, known rates of hydrogen and oxygen (in the stoichiometric ratio) were introduced directly into the space above the reactor solution. The three catalyst bed temperatures (inlet, center of bed and outlet) and the temperature rise of the sweep gas were recorded at equilibrium * for each hydrogen flow rate. From this a calibration curve correlating temperature with hydrogen flow was constructed (Fig. 12).
2. Keeping the recombiner system at the same conditions, the reactor power was increased stepwise and the equilibrium temperatures for each power level were recorded in the same manner. The plot of these data is shown in Fig. 13.
3. Using any one of the four temperatures as a parameter, the rate of hydrogen formed radiolytically was correlated with reactor power level (Fig. 14). Good agreement was obtained using any of the three catalyst bed temperatures, or the sweep gas temperature rise. Prior to installation of the recombiner system, the rate of water addition to keep the reactor at constant volume was 0.49 mole/kw-hr. The rate of H_2 evolution as determined above was 0.55 mole/kw-hr. As shown in Fig. 13, the catalyst chamber temperatures provide a reasonably good method of measuring reactor power. Seemingly, the temperature of the inlet region of catalyst bed would provide the best reference since it is the least dependent on the surroundings. However, if the catalyst should become poisoned, the main reaction zone would move deeper into the bed and result in lower reference temperatures. Consequently, the most desirable reference is the sweep gas temperature rise across the chamber at a given total rate of flow of air.

D. Conclusions. The recombiner system has been operated for the past nine months (approximately 16,000 kw-hr). Neither the standby catalyst chamber nor the external heaters have been used to date. With the exception of the blower seal, the system has worked very satisfactorily and no other changes are contemplated.

* Complete temperature equilibrium for a given recombination rate is not established even after many hours of operation. Since only slow constant drifts occur after the first 40 min, data taken after this time were used in the experiments.

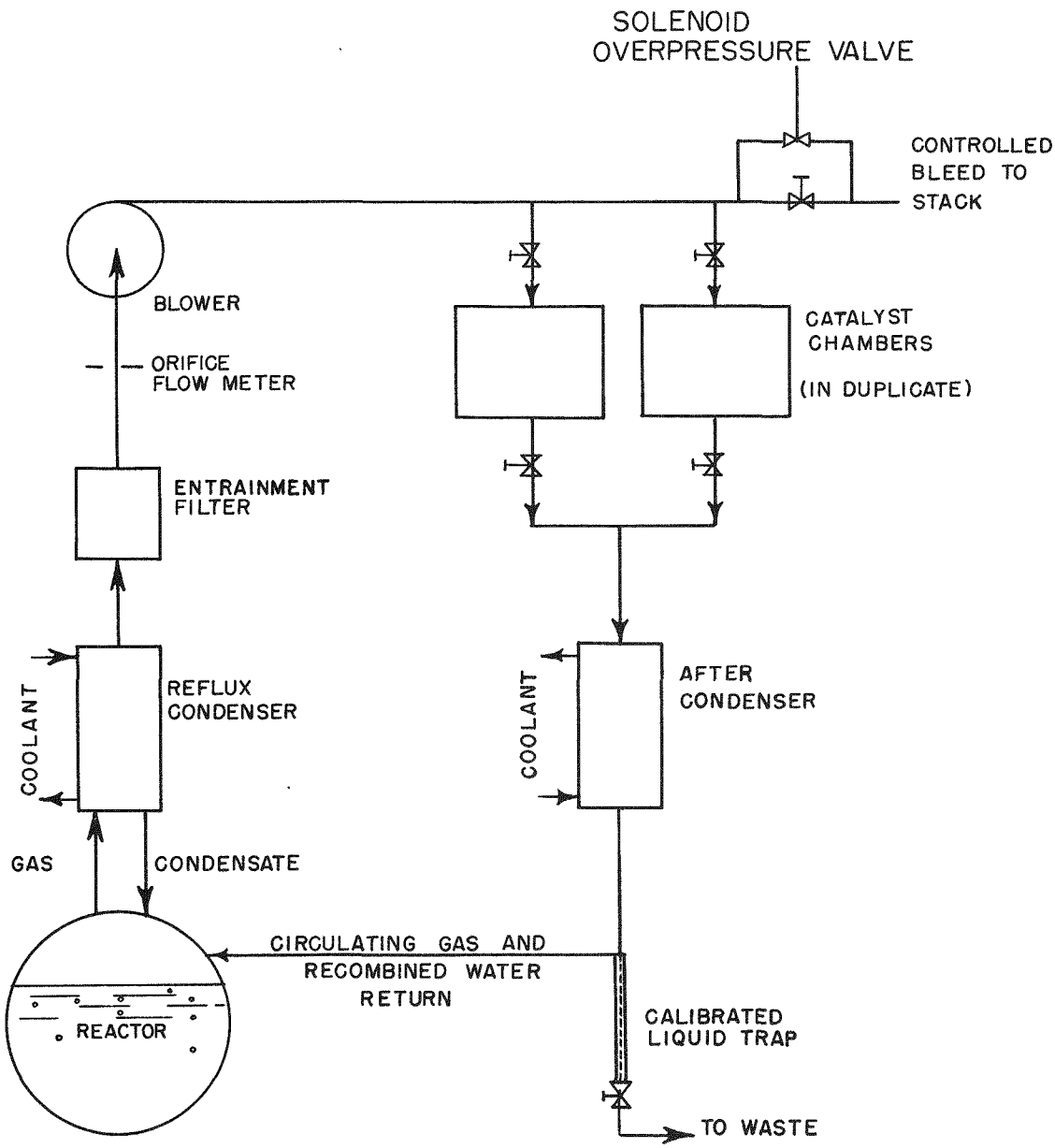
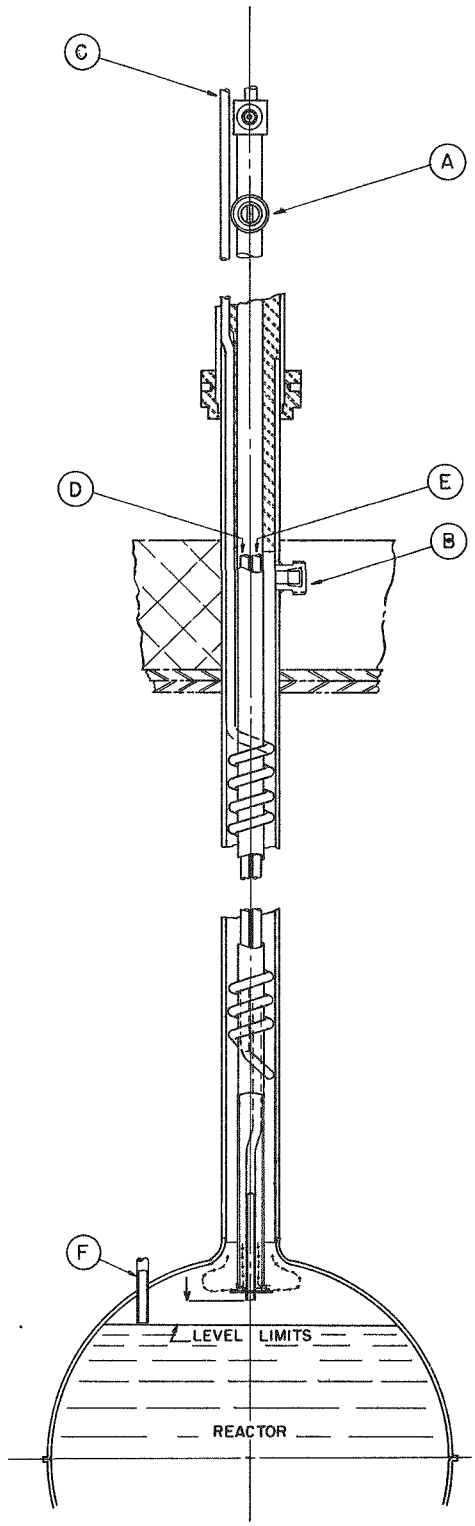


Fig. 1. Block diagram of recombiner flow system.



A. Gas return to reactor
B. Gas exhaust to catalyst chambers
C. Coolant inlet
D. Coolant outlet
E. Upper liquid level tube
F. Lower liquid level tube

Fig. 2. Reflux condenser.

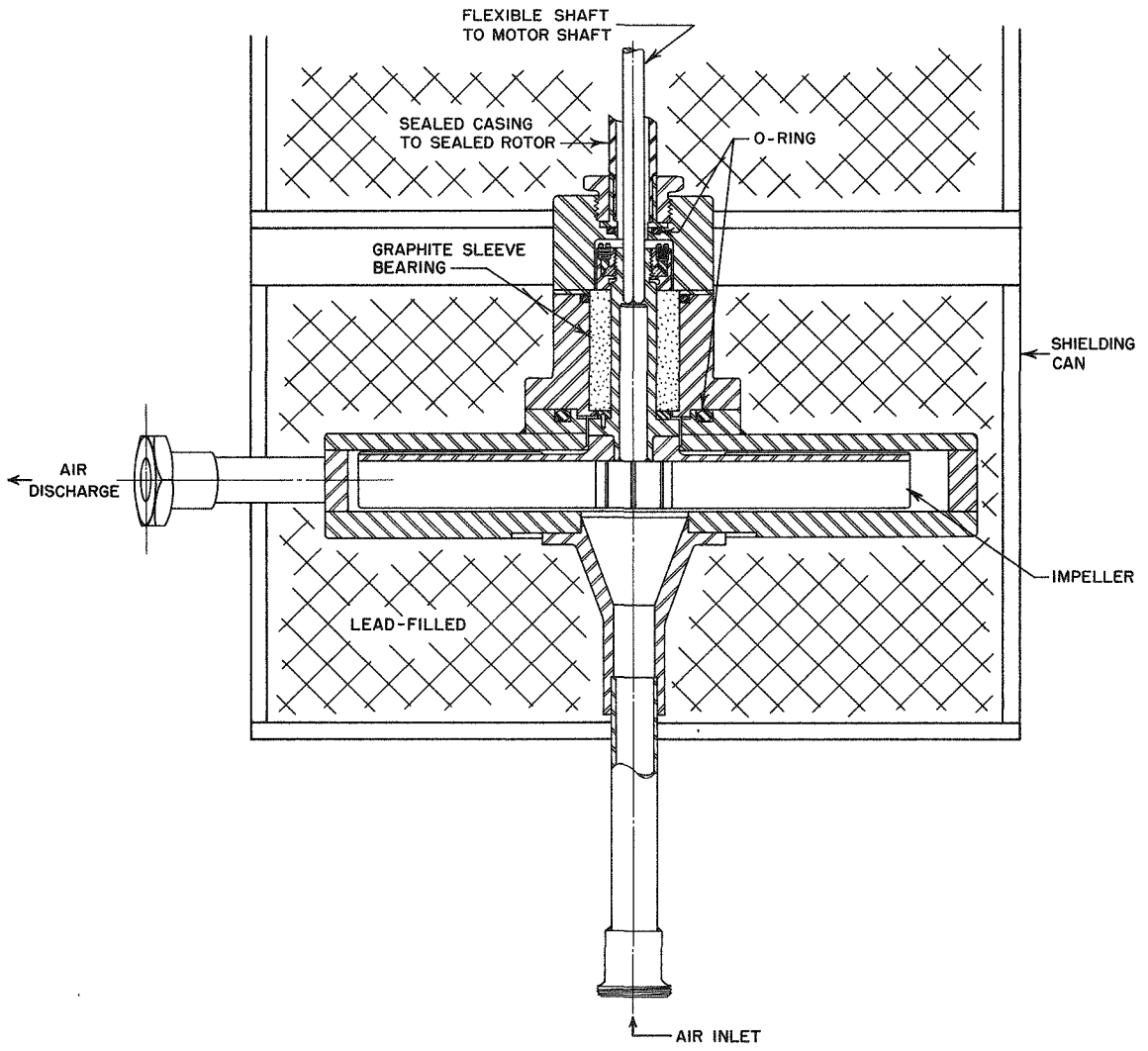


Fig. 3. Blower assembly.

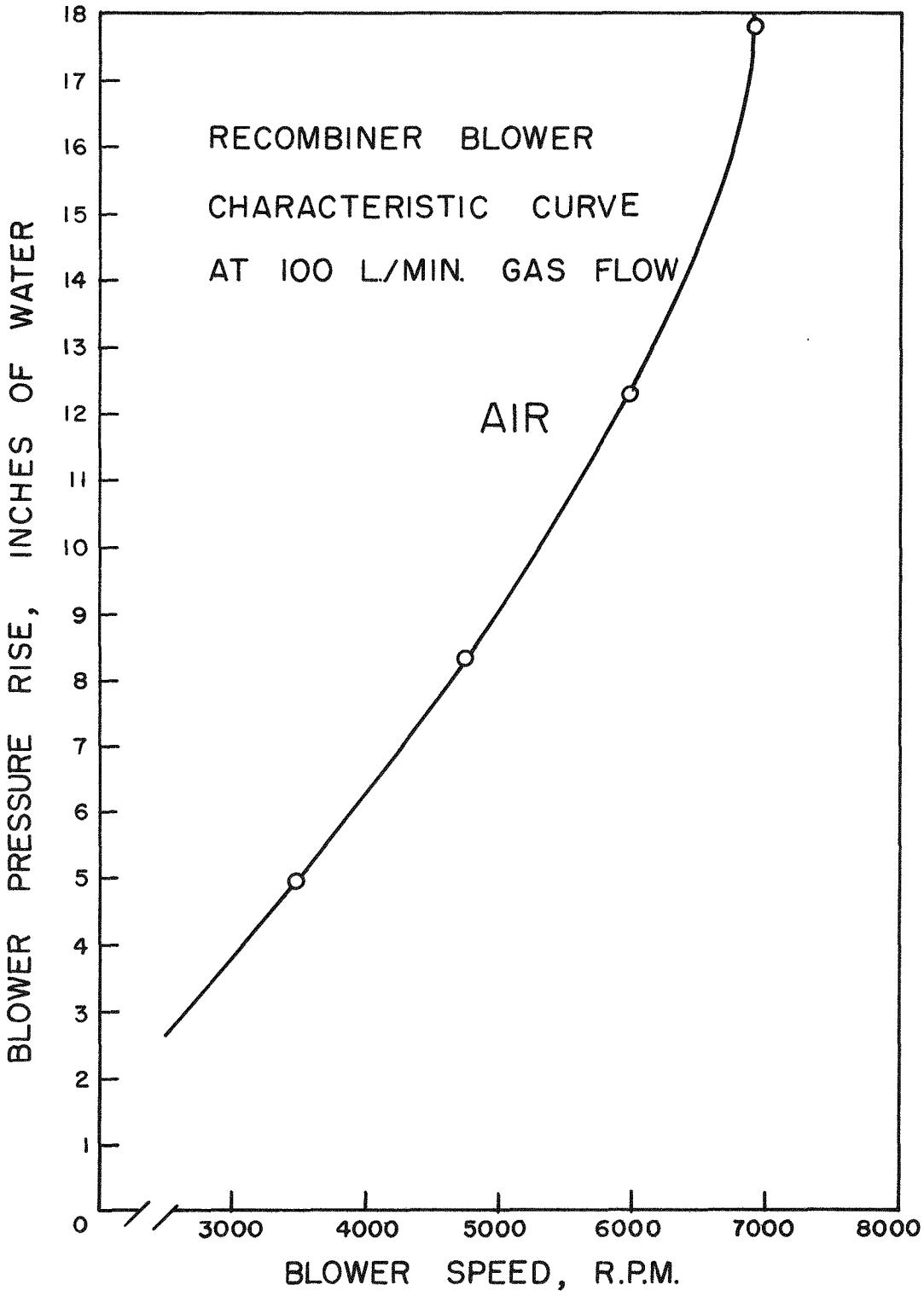


Fig. 4. Blower performance curve.

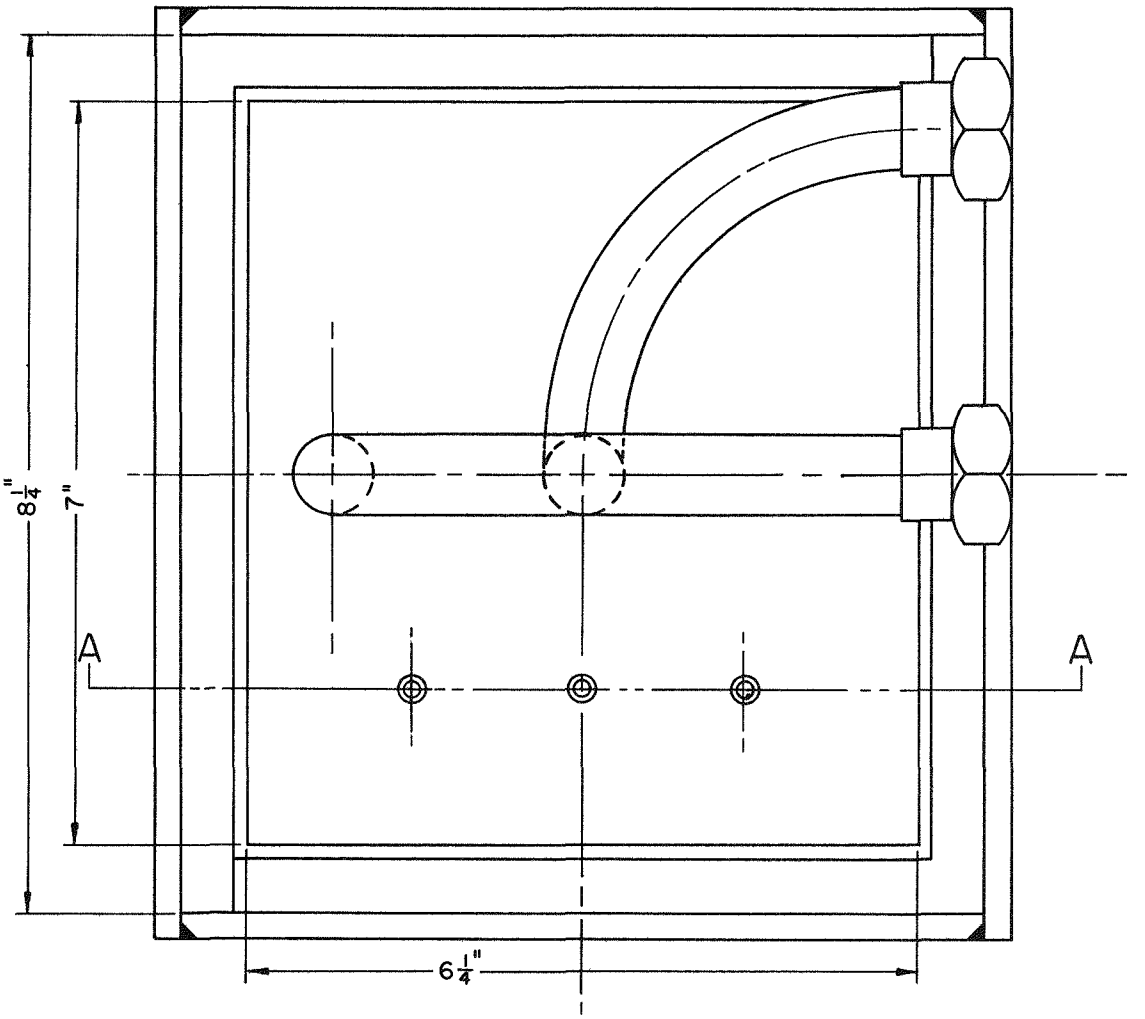


Fig. 5. Catalyst chamber.

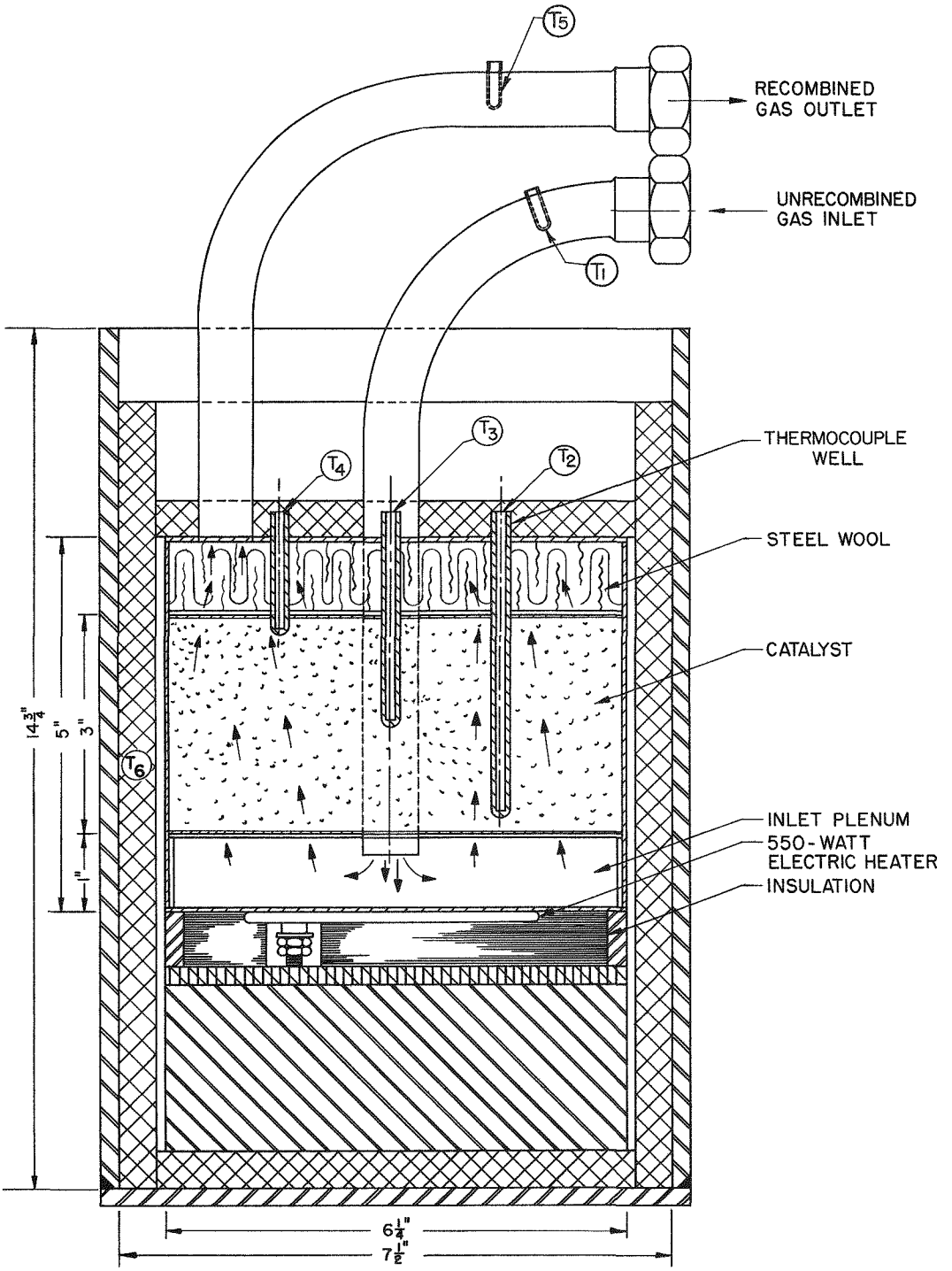


Fig. 5A. Section A-A of Fig. 5.

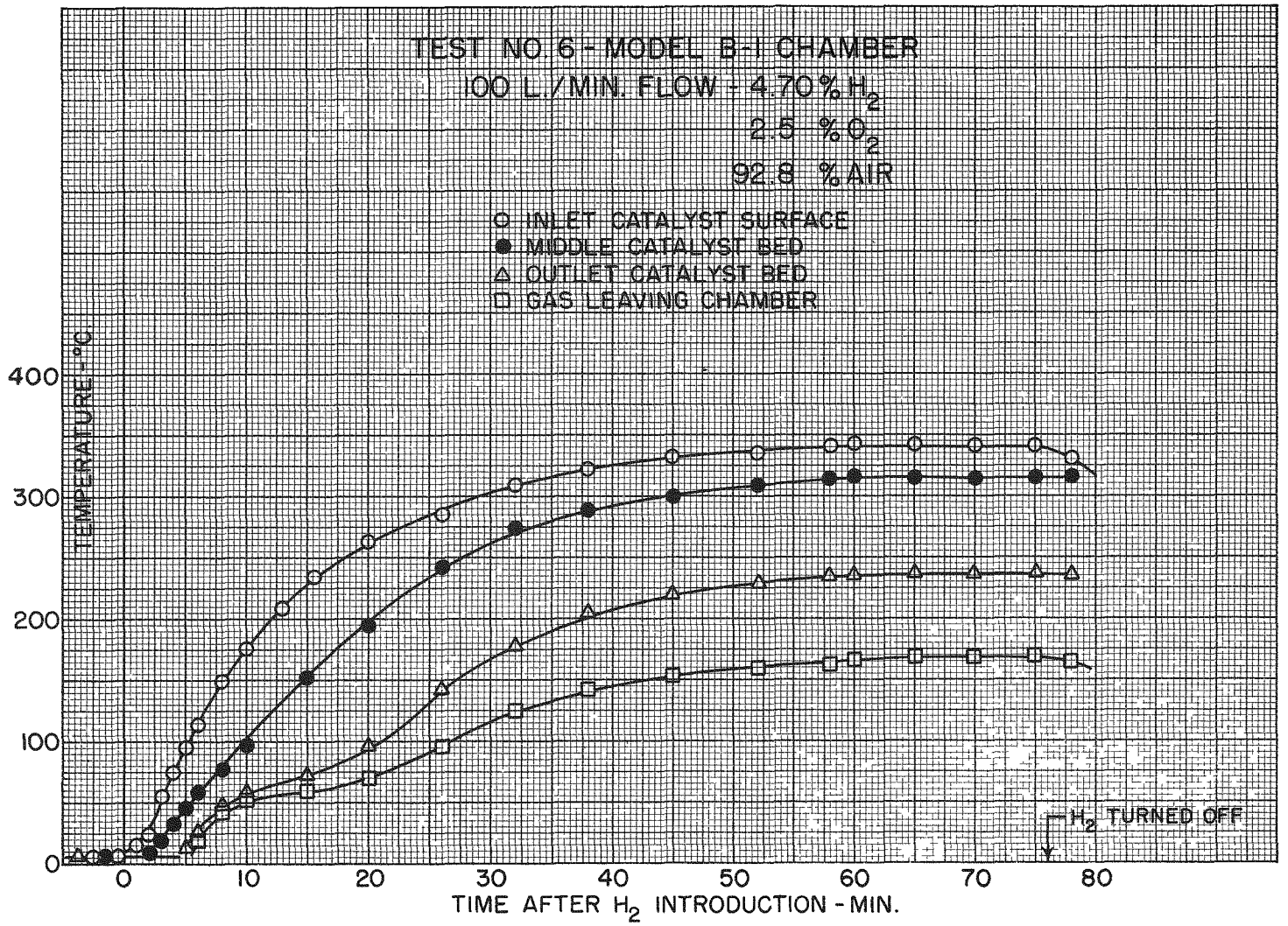


Fig. 6. Catalyst chamber test curves.

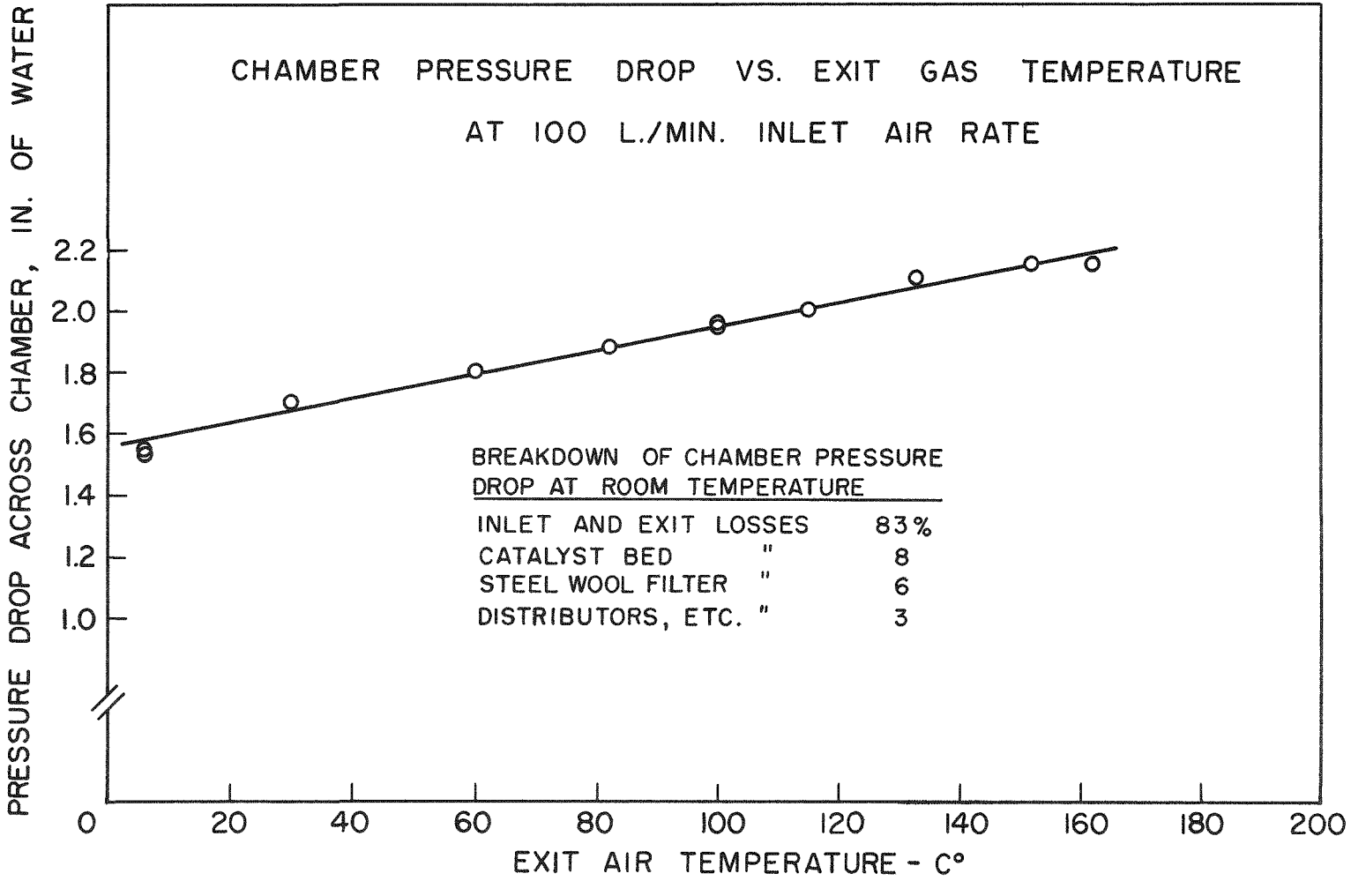


Fig. 7. Effect of gas temperature on catalyst chamber pressure drop.

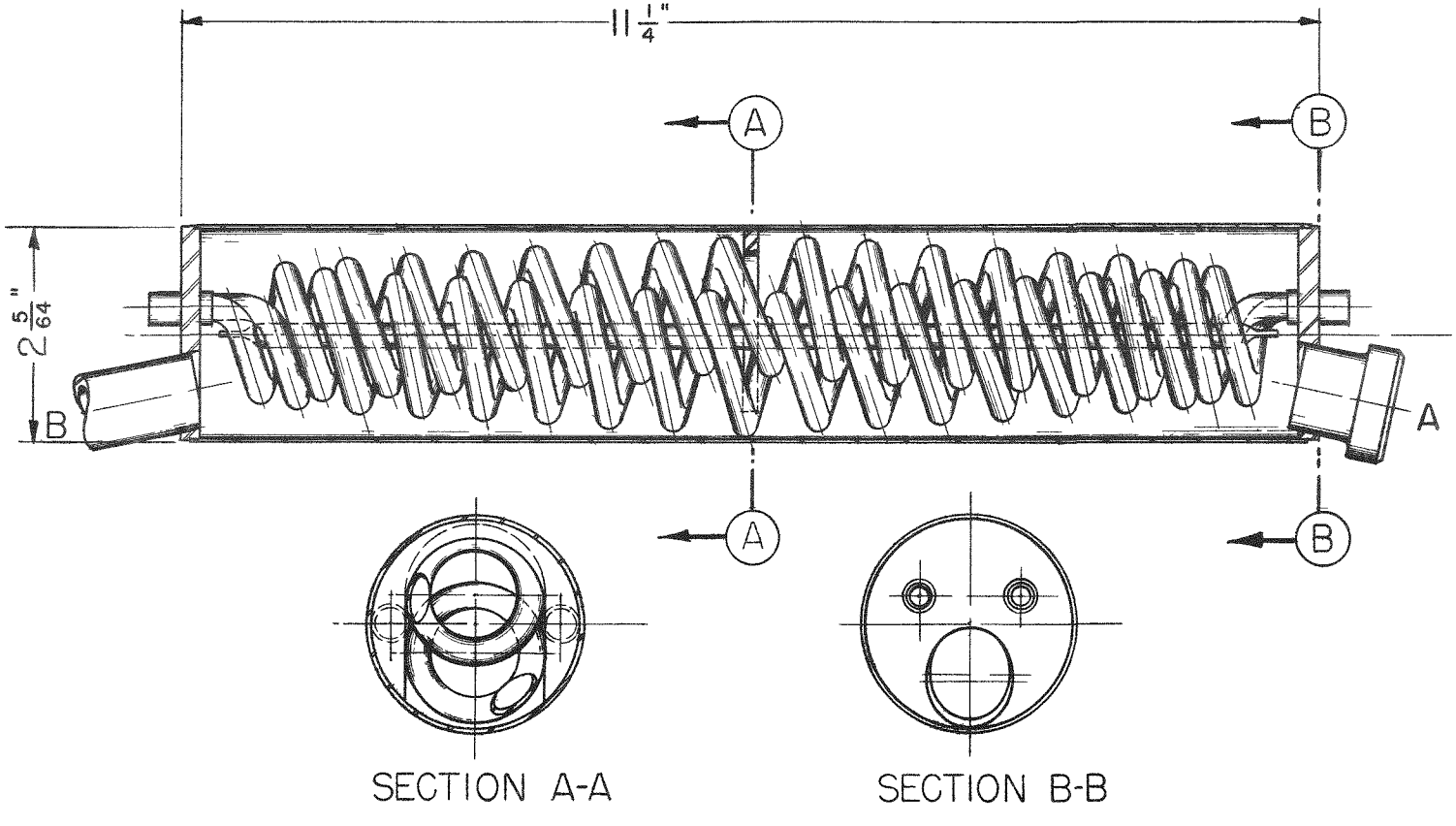


Fig. 8. After condenser.

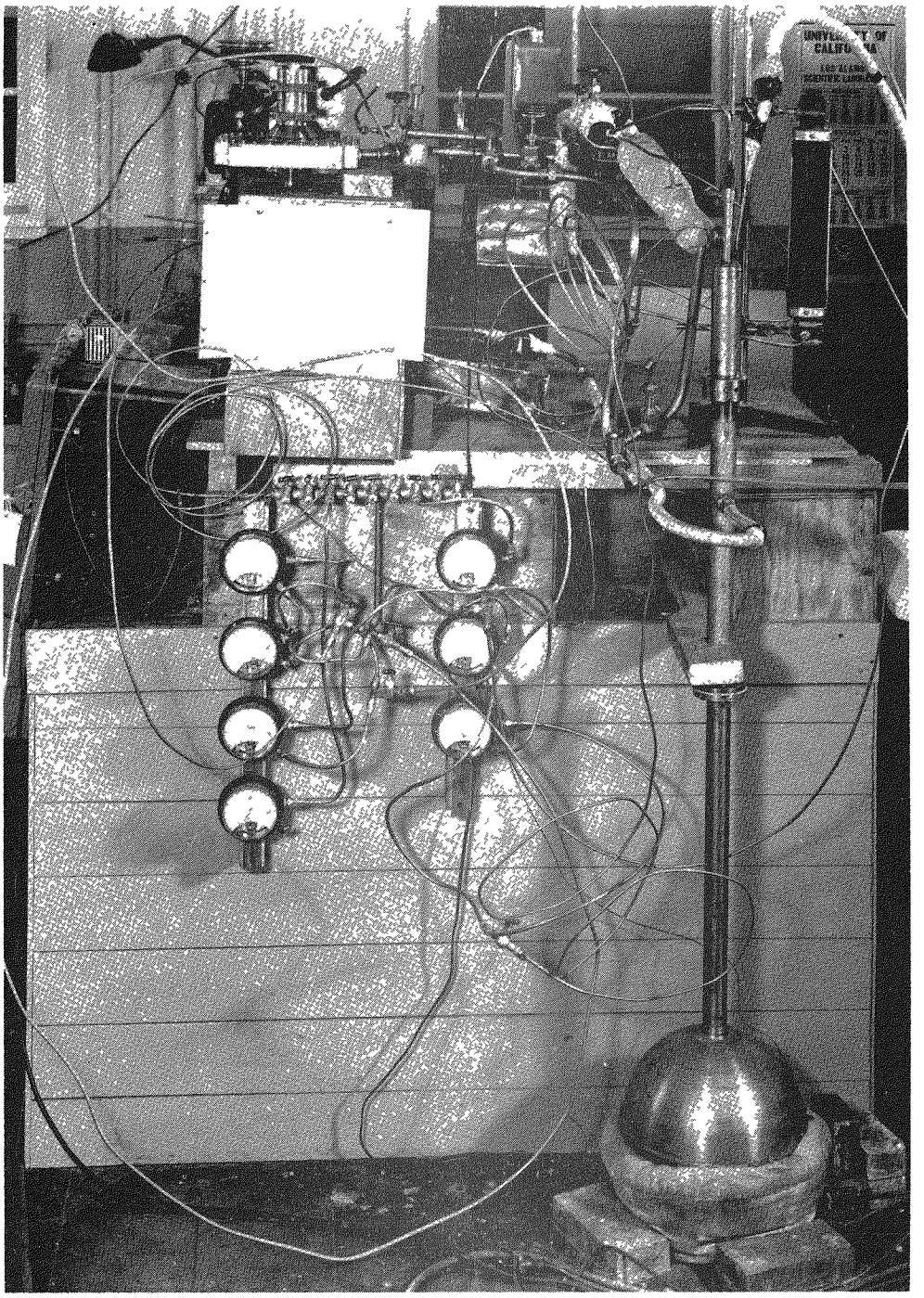


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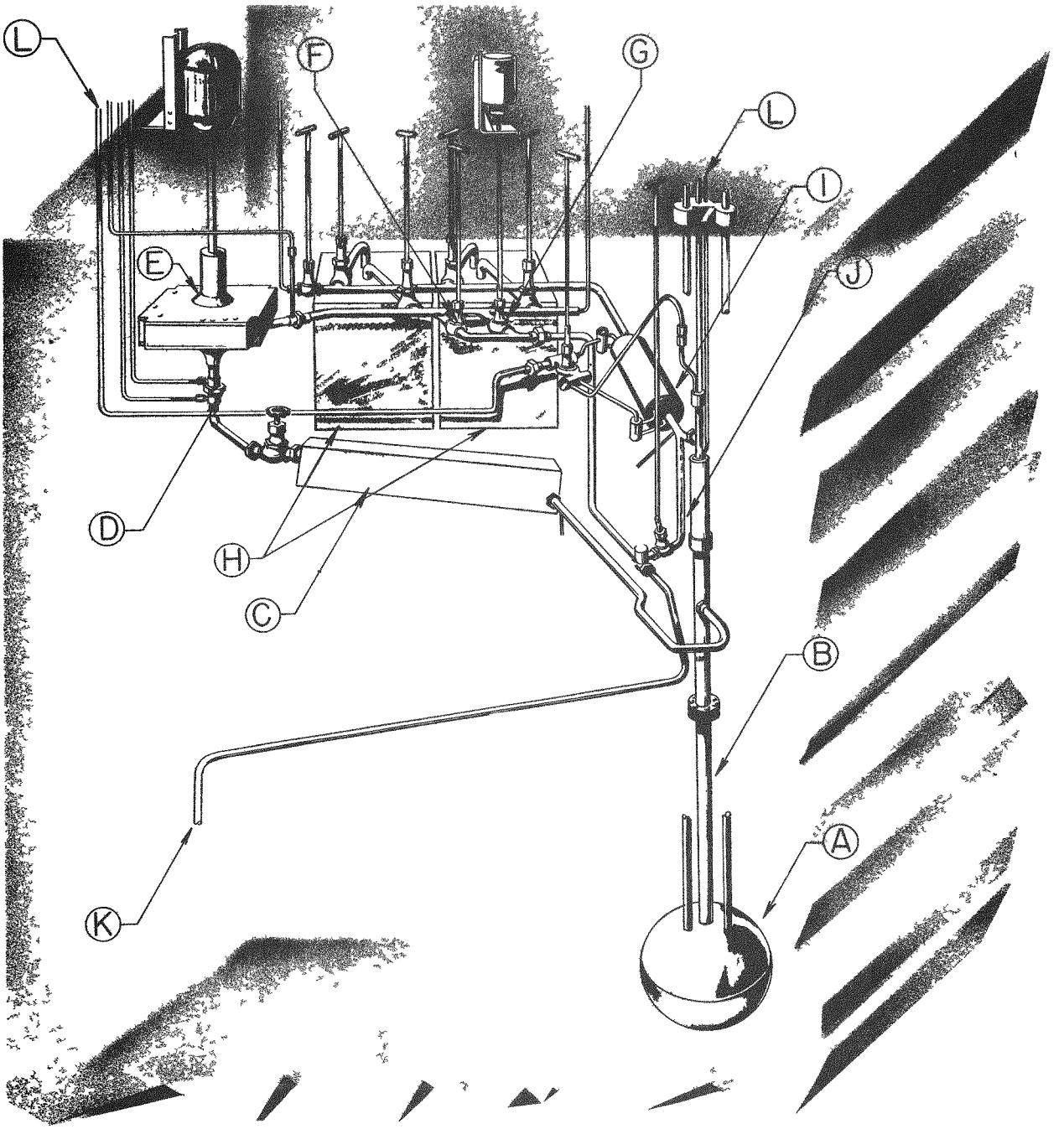
Fig. 9. Mock-up assembly.



A.	Reactor	E	Blower	I.	After condenser
B.	Reflux condenser	F	Control bleed valve	J.	Liquid trap
C.	Filter	G	Overpressure valve	K.	To stack
D.	Orifice meter	H	Catalyst chambers	L.	Coolant

Fig 10 Installation phantom view

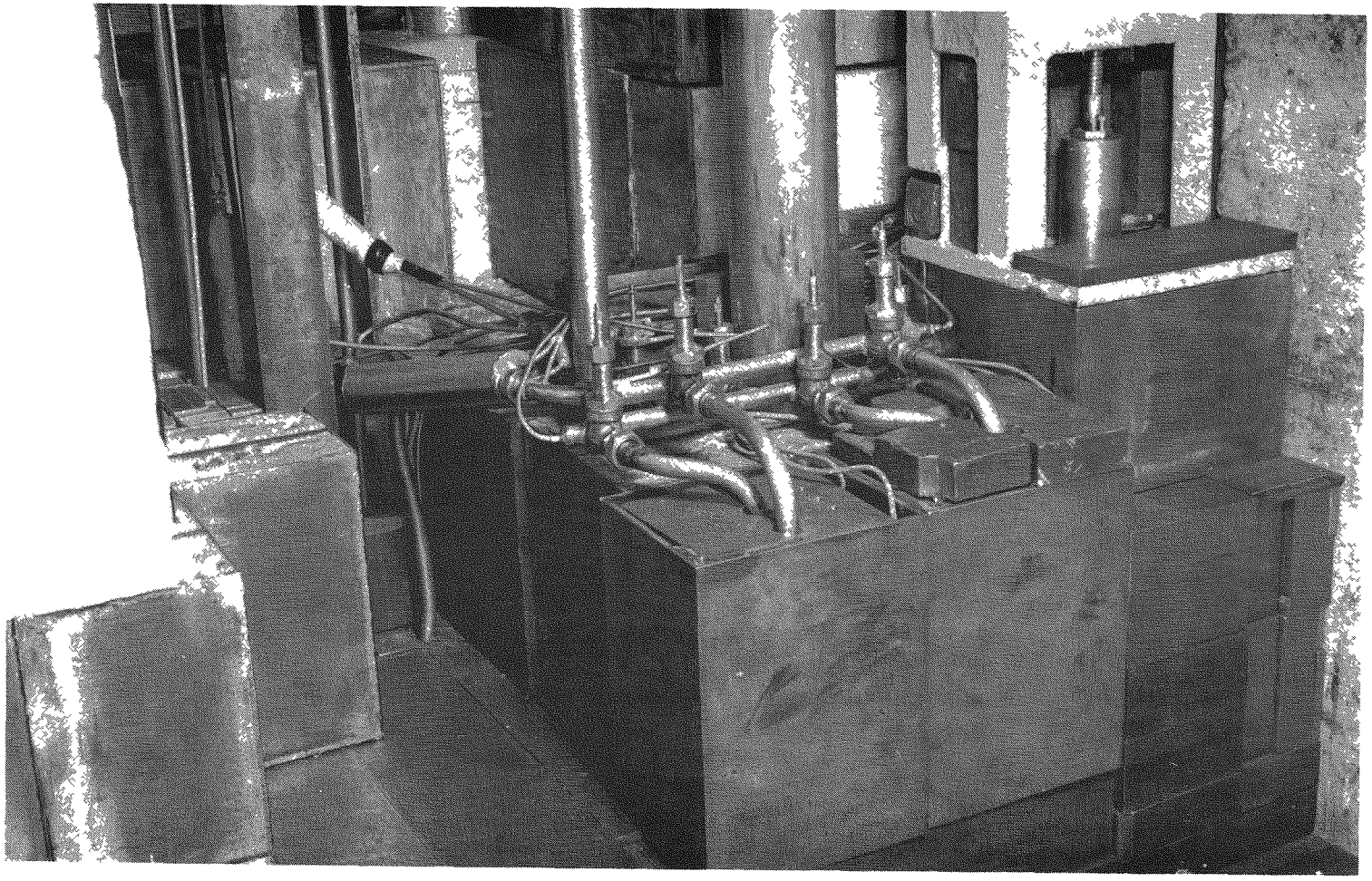


Fig. 11. Photograph taken during installation.

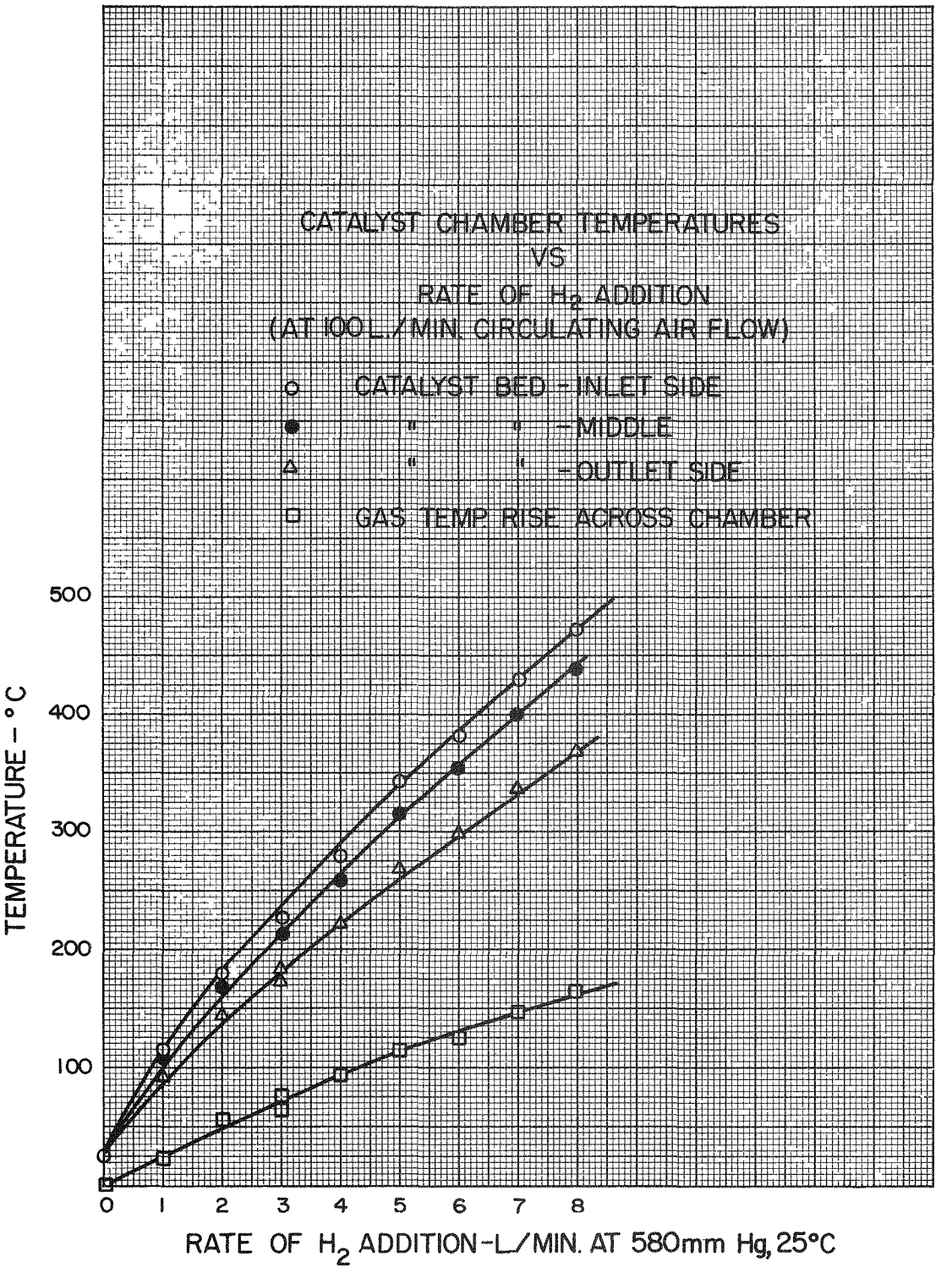


Fig. 12. Plot of catalyst chamber temperatures as a function of hydrogen flow rate.

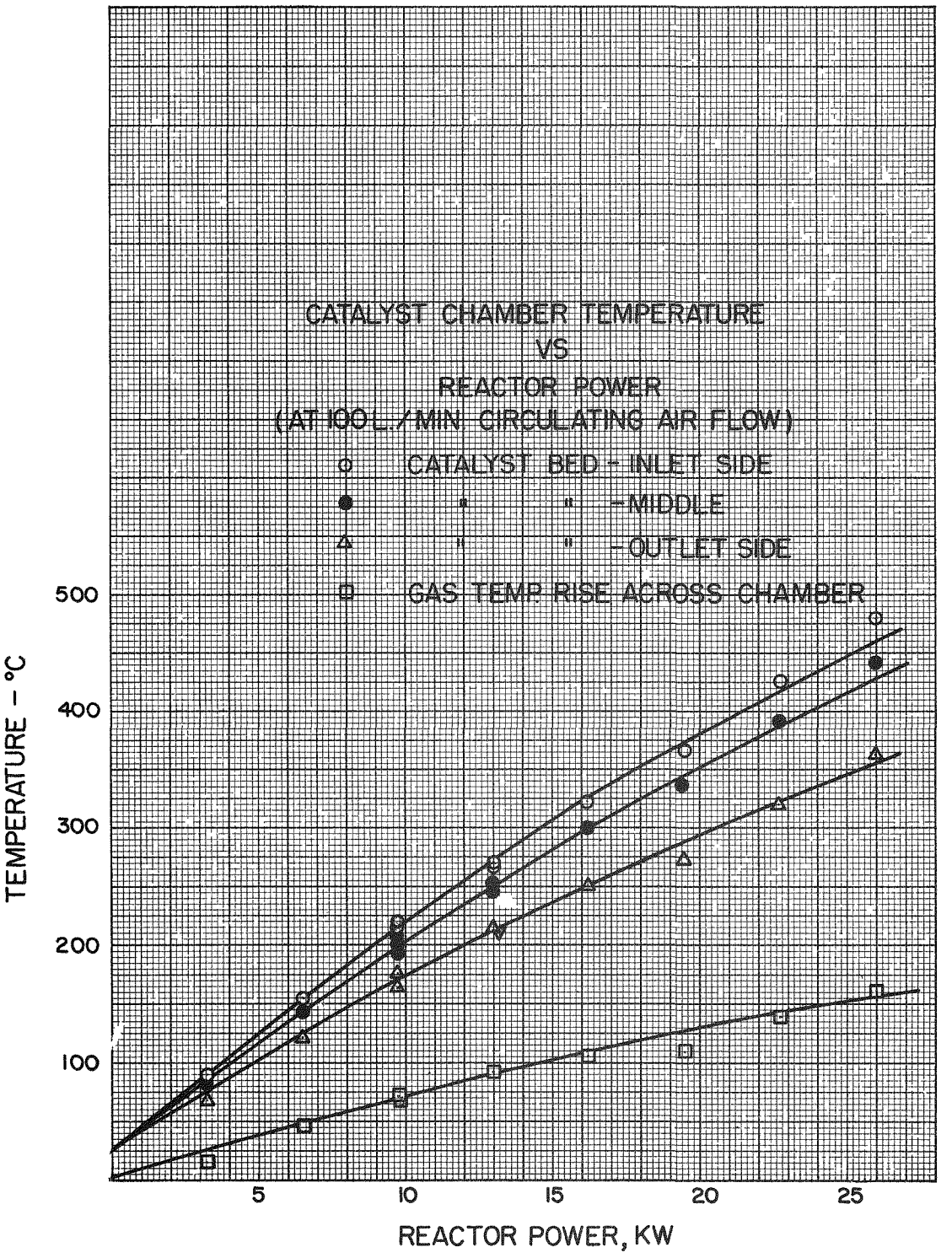


Fig. 13. Plot of catalyst chamber temperatures as a function of reactor power.

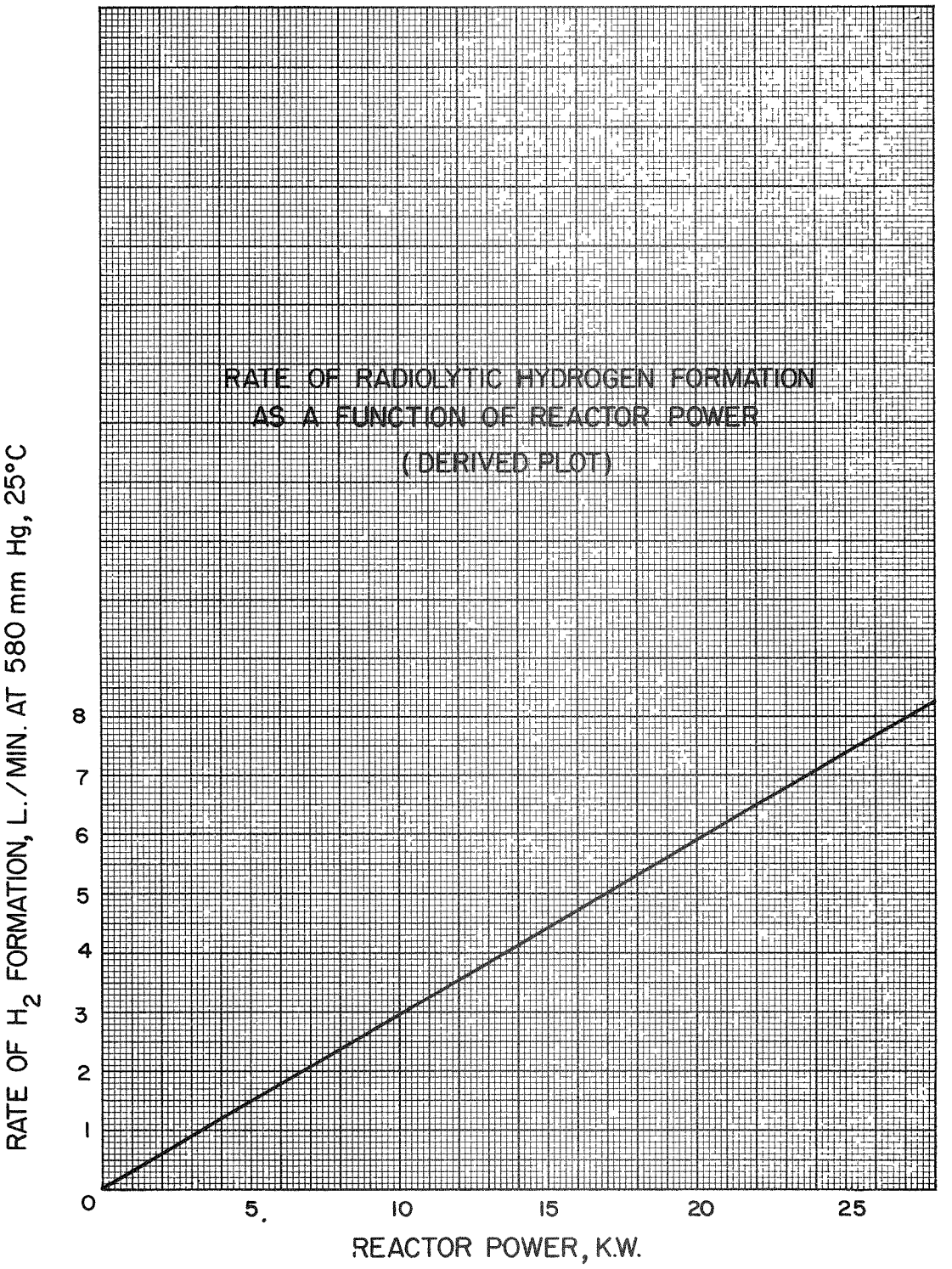


Fig. 14. Plot of rate of radiolytic hydrogen formation as a function of reactor power.