

nitrogen jacket during pressurization and depressurization deformed and cracked a stainless-steel expansion bellows on the steam-water line from the water jacket. This bellows, contrary to the understanding of HYGAS personnel, had been designed for 150-psi internal pressure and only for 40 to 50 psi external pressure. The pressurizing procedure was modified to prevent a similar occurrence.

The refractory grid in the second stage was in good condition, except that the center plug had fallen through to the lower heat-exchange bed sometime during the cooldown period. The center plug showed no sign of corrosion or erosion. The opening in the grid into which the center plug fits was measured and it was learned that a poor fitting job had probably caused it to fall out. A refractory installer refitted the outer course of the refractory deck; however, the new center opening for the plug had no taper. Rather than risk the loss of another center plug, a taper was ground in the hole. This work proceeded slowly because the grid was made of very hard, high-alumina refractory. After the plug for the refractory grid was fitted, the patching in the spalled section above the refractory grid was also completed. The refractory in the hydrogasifier vessel was cured at 600°F for 2 days, after which a starting bed to the reactor was charged and the temperature of the reactor was brought up with the start-up ring burner. The spalling on the wall all occurred in the area of the fluidized bed, where coal and sand fill any cracks that are formed during the heating and cooling. The patch refractory used was a ramming mix which has good wear and heat qualities. The freeboard area of the fluid bed was in good condition.

All of the valve seats on the Wilson-Snyder pumps became swollen when in contact with toluene. The polyurethane and Buna-N seats were not able to withstand continuous operation. Both Teflon and Viton seats were tested; the latter was believed to be durable even when in contact with toluene. The seal rings on the slurry circulation pumps were also swollen, and were replaced. The impeller on the circulation pump was eroded, although it had been coated with an erosion-resistant Noroc coating, which had been used successfully in slurry pumps for circulating oil-catalyst mixtures in many refineries.

Ingersoll-Rand felt that the failure of the Noroc coating had most likely been caused by an inadequate application of the coating to the base material. They suggested two wet ends: One would be coated with tungsten carbide and the other with an improved ceramic material. Meanwhile, a slurry circulating pump was reassembled to perform wear tests on the Wilson-Snyder pumps. One of the Wilson-Snyder pumps had been installed with a set of Viton seats; the other pump had a set of hardened Type-410 stainless-steel seats. After installing two letdown chokes in the discharge line from the pump back to the suction, circulating tests were run to see if these two materials could solve the wear problem.

The new tungsten-carbide-coated wet end was installed on a slurry circulating pump. This pump was opened after 5.5 days of operation. The impeller was worn but somewhat less than the Noroc-coated impeller; however, there was still too much wear to be acceptable. Also, the stuffing box extension and the part of the casing that fits over it were gouged as in the previous pump problem. It appeared that putting on hard surface coatings was not the right answer to the erosion problem.

This problem was discussed with many companies, among them the Black Mesa Pipeline Company, Consolidation Coal's pipeline staff and its Cresap staff, the C. F. Braun and Company pump personnel, and, of course, Ingersoll-Rand. Through these contacts a number of manufacturers of low-pressure, sand-and-gravel slurry pumps were consulted, including the Wilfley Company; Barrett, Haentjens and Company; and Wilson-Snyder Company. The consensus was that, although high-speed pumps, such as the 3500 rpm pump then used at the pilot plant, could be made to work on slurry, it would be much better to try to operate slurry pumps at lower speeds. All other factors being equal, wear increases rapidly with speed. Speeds within the range of 1150 to 1750 rpm were then suggested.

Two 1750-rpm motors were installed on the slurry circulating pump, one in the low-pressure coal-toluene slurry system and the other in the high-pressure char-water slurry system. The reduced pressure head from 1750-rpm operation appeared to be tolerable in the char-water slurry system. Engineers agreed that, if the wear at reduced speed were acceptable following test No. 5, HYGAS personnel would consider this a permanent solution, because there were no slurry-pump manufacturers who could supply a pump for high-pressure operation. Ingersoll-Rand returned a tungsten-carbide-coated high-pressure slurry pump which was installed to replace the pump with the worn Noroc coating.

On the low-pressure system, the reduced head probably was marginal for operation. Slurry circulation could be maintained by removing the choke on the pump discharge and by using the largest allowable impeller. At the same time, quotes were reviewed for a slurry pump of different make. For back-up purposes, the speed of the steam-turbine-driven slurry pumps was reduced to 1750 rpm. If the only problem with the Ingersoll Rand pumps was their high speed, it was reasoned that - by reducing the speed - the pumps might be used for future tests without further modifications.

The Type-410 stainless-steel valves and seats on one of the Wilson-Snyder mud pumps were examined after 60 hours of operation. The seats were slightly pitted, but not by an inordinate amount. A new washer was welded to these valves to improve valve centering. One of the three cylinders was not pumping during the test, and it was suspected that the valve got caught in its travel and did not seat properly. Viton seats were also examined after 48 hours of testing. They were only slightly swollen by the toluene at 145°F and were in much better shape than the previous elastomer seats. This pump operated satisfactorily throughout the test. Wilson-Snyder examined the valve seats and molded other sets of Viton seats which were harder than the ones used to that time.

For slurry feed, the Wilson-Snyder high-pressure mud pump was now used with Type-410 stainless-steel valves. Up to this time, operators had been able to run the plant without any loss in pressure or flow.

When the discharge slurry pump was opened, excessive spalling and wear were found in the Noroc coating on the impeller and the casing. A spare pump was shipped to have it coated with tungsten carbide. The quench water circulating pump impeller was found to be badly corroded. During

the test, efforts had been made to maintain a pH of 8 in the quench water by adding an ammonia-water solution through a metering pump. The piping was now changed to inject anhydrous ammonia directly.

The new control on the coal mill operated satisfactorily; the dust collector could now be heated to over 200°F before crushing began. Based on experience gained by the Rapid City group, three of the six journals in the mill were removed in an effort to obtain a better size distribution.

The hydrogen compressor was disassembled. On one discharge valve, a valve guide made of higher nickel-content steel had cracked. The third-stage rings were also found to be badly worn and leaking. The rings and the rod were replaced before the next test. Graphite-filled Teflon rings, which had worked well in the first two stages, were used in place of the bronze-filled Teflon rings. The filter arrangement on the suction of the compressor was also improved.

Next, the hydrogen compressor developed a serious flaw: The liner inside the third-stage cylinder was bowed 0.02-inch in diameter, while the tolerance was only 0.002 inch. As a result, the piston would not slide freely even when cold, and certainly would not slide freely when heated. The third stage was completely dismantled and shipped to Ingersoll-Rand's Painted Post plant, where the flawed liner was cut out and a new one installed. This was believed to have been the reason for difficulties with the third stage since start-up operations had been initiated.

Field alignment of this cylinder revealed that it was far out of line. The cylinder connections to the distance piece and from the distance piece to the cross-head section had been machined with rabbet fits, which should have been self-aligning. Even though this machine was supposed to have been aligned by Ingersoll-Rand in 1970, and any subsequent disassembly and reassembly should not have disturbed the alignment, the fact that the misalignment had become so great seemed to indicate that the problem had occurred much earlier than the time it was detected.

The third stage was aligned by Mr. Harold Cleveland of Ingersoll-Rand's Painted Post plant, an expert on wire alignment. The new liner was true and in alignment. The overall result was that the 0.046-inch steel shims, which were installed between the cylinder pedestal and a concrete support when the machine was originally assembled by Procon, were removed. In any event, the cylinder was aligned without need for any machining.

After the alignment and reassembly, the compressor was run for 75 hours when the first-stage discharge valves were found to be worn on the end channels. Ingersoll-Rand recommended that some of the metal be ground off these end channels to avoid having the channels come in contact with the metal before the valves are fully seated. This was done on all the suction and discharge valves for the first and second stages, and the compressor was put back in operation. All outward indicators were within design limits after two days of operation, including discharge pressure, temperature, and temperature rise from the coolant. The performance of this compressor was watched carefully in hopes that the valve problems, which had plagued the plant, were solved.

A criterion was finally received from Ingersoll-Rand for controlling coolant flow through the third stage. The temperature rise in the coolant had to be maintained at a maximum of 10°F under any operating conditions to prevent excessive thermal stress on the compressor.

More crushing tests were performed on the coal mill. The initial test showed that the moisture could be reduced from the previous level of 18% down to about 7 to 8%. In addition, the coal particle size distribution was much better than before, with a -100 mesh fraction of around 12% instead of the previous 25 to 30%. Minor adjustments were made on the coal mill, particularly to provide inert gas purging when the mill shut off. This was to prevent oxygen build-up that could cause burning in the dust collector. Some of the earlier problems encountered in start-up operations were attributed to air leakage into the coal mill through the windbox where the heating caused the metal to buckle. These areas had been patched and the controls were improved.

3.5.9 Gasification Test No. 5, April, 1972

The fifth gasification test was started on April 19, and was terminated April 28, 10 days after the start of coal feeding to the reactor. During this period, 42 tons of lignite were added at a feed rate of approximately 1.5 tons per hour. Feeding was intermittent because of problems encountered primarily with the quench separator system and the slurry dryer stage in the reactor. Operation was at 500 psi.

The slurry dryer malfunctioned almost from the beginning of the test. The radiation gage and the differential pressure measurement both showed that the bed level did not respond to coal feed as it should have. Although coal could be transferred from the slurry dryer through the first and second gasifier stages, through the heat-exchange bed, and all the way out to the slurry discharge system, such transfer could not be maintained on a steady basis for extended periods of time because solids flow could not be maintained out of the slurry dryer bed. However, automatic control of solids flow was achieved between the two stages of the reactor.

Because the slurry dryer was not functioning properly, large amounts of solids were carried over from the reactor into the quench system. Coal mixed with water and toluene created a suspension and perhaps an emulsion in the quench systems, which directly affected the operation of the interface controller in the quench separator. As a result, the proper interface could not be detected or maintained, and large quantities of toluene were lost from the system. The large amount of coal in the quench system clogged the filters and prevented the feeding of clean recycle toluene for seal flushes. This clogging, coupled with a hydrogen compressor problem discussed below, caused the termination of the test. During the test, hydrogen flow to the reactor was between 50,000 and 60,000 standard cubic feet per hour and steam flow was 1000 pounds per hour.

When the reactor was cooled and opened for inspection, the slurry dryer bed was packed with wet solids. Water had dripped in through two leaky valves upstream of a spray nozzle used for temperature control. The top reactor cover was removed and the wet solids were cleaned out of the slurry dryer bed. The leaky block valve and control valve for the spray nozzle were repaired.

The slurry dryer grid was in good condition: The new high-temperature sealant developed by HYGAS researchers appeared to have held well during the test. There was no sign of bypassing around the grid and no evidence of erosion caused by bypassing near the grid area. Several areas in the slurry dryer bed were modified before beginning the next test; for example, the slurry inlet nozzle was lowered and one more pressure tap and a stilling well were added on the withdrawal pipe.

A rupture was detected on the internal piping in the nitrogen jacket during the reactor cooldown period. The expansion joint on the lift-line in the nitrogen jacket area was ruptured at a weld made between Incoloy Type 800 and Type 446 stainless steel. The expansion joint was made with a 0.5-inch free play in the ends of the tie rods when the lift line was cold; when hot, the expansion joint contracted. From the inward bending of the two end plates on the expansion joint, it was apparent that pipe shrinkage during cooling had created a smaller pipe size than when it had been cold; as a result, the tie rods were restrained by the end plates, causing the eventual rupture at the weld. A spare expansion joint was installed in preparation for the next test.

While reassembling the internals, a second rupture was found below the expansion joint of the feed line to the mixing pot of the first-stage lift reactor. Again, the joint failed from tension and was separated from its original position by approximately 1 inch. The rupture was in the parent metal, not in the weld. That section was repaired and an extra 1 inch of allowable expansion was added in the bellows to relieve cooling stress. Although the original allowance was 0.5 inch, which should have been more than adequate, 1.5 inches was then available to accommodate expansion. The second-stage gasifier was then cleaned. In addition to coal, it contained a large amount of clinkers, which apparently formed from the combustion of hydrogen and the high-pressure air used to maintain the temperature during the test. The grid and refractory balls used to prevent weeping on the grid were not attacked. All of the clinkers were above the air injection point. The clinkers and ash from the lignite both showed complete fluidity at 2200°F and incipient fusion at around 2000°F. This is a low ash fusion temperature, and undoubtedly was the cause of the clinker formation in the second-stage gasifier. With this particular lignite, combustion must occur below the second-stage refractory grid to avoid any hot spots in the bed that could cause the lignite ash to soften and fuse.

Plans were made to inject air into the then existing natural gas-air burner ring, to use the existing spark igniter for ignition, and to burn air with hydrogen for heat-up (instead of natural gas). The monitoring was done through a television transmitter in the reactor. Various hydrogen and air flow rates were tried without initial success. After several modifications, combustion could be sustained under certain hydrogen-to-air ratios. The range of combustion was broadened to permit reliance on the hydrogen-air combustion for the reactor heat-up. The difficulty in burning the two gases resulted from the great difference in their molecular weights: The air mixes too rapidly with the hydrogen, resulting in a mixture which is above the upper combustion limit of hydrogen in air. Several means were tested to reduce the mixing and to allow the attachment of the flame to the refractory wall, which would eventually act as an anchor for the flame.

The slurry circulating pump (which operated at 1750 rpm) was opened after the test. The casing and the stuffing box extension were essentially not eroded. The impeller was badly worn, however, and had to be replaced. The spare Ingersoll-Rand circulating pump, with steam-turbine drive and with its internal parts coated with Garlock 202 ceramic coating, had been on circulation test for 8 days. It was also worn and was no better than the tungsten-carbide-coated pump. The Wilfley slurry circulating pump was installed and tested. It pumped smoothly and delivered the proper amount of head on toluene.

In the high-pressure slurry circulating pump, the tungsten carbide coating on the stuffing box extension was not attacked. The impeller was worn, but not nearly as badly as before; however, the coating on the casing was peeled. After a discussion with Garlock and Ingersoll-Rand's representative, an improved version of tungsten carbide coating was applied to another casing and impeller. A tungsten-carbide-coated Ingersoll-Rand pump with nickel chrome substrate was installed for the high-pressure slurry circulation.

The Wilson-Snyder mud pumps operated well through the whole period of Test No. 5. The Type 410 stainless steel valves, examined after three days of operation, were only slightly pitted. The valves were reinstalled; operation continued with the same set of valves to the end of the test without any loss of pressure or capacity. The valves were reexamined after the test; they were still only slightly pitted and could be used again. The Viton valves, having a 95 durometer reading, were installed. The metal valves appeared to be a good candidate as a solution to the wear problem.

The hydrogen compressor, which had been operating for 12 days during Test No. 5, operated well up until the last 2 days. At that time, discharge temperatures and pressures began to rise, indicating problems with the compressor. Ingersoll-Rand's calculations showed that the discharge temperature for the third stage was 90°F higher than the theoretical gas temperature based on the measured compression ratio. Carbon-like fine deposits had collected on the suction filter and had created a pressure drop of more than 2 psi; this indicated that the rings in the compressor were worn.

The hydrogen compressor was disassembled in the presence of an Ingersoll-Rand engineer and Mr. Trevithick of C. F. Braun, who visited the HYGAS site at the request of OCR. Meetings were also held with Mr. Feldmann, of the Ingersoll-Rand Painted Post plant, and Mr. Paul Towson of OCR. After examining all of the parts, the Ingersoll-Rand engineer concluded that the reason for the excessive compression-ring wear on the second and third stages was the ring design. He believed that a new ring design, incorporating overlapping stepped joints on the rings should prevent gas from leaking preferentially, through the opening in the ring, compared to the solid portion of the ring. Cook fabricated these on a "crash basis".

3.5.10 Gasification Test No. 6, June, 1972

After modification of the ring burner, the combustion of hydrogen with air was achieved successfully under a wide range of heat inputs. The combustion was complete, smooth, and immediate upon air introduction. Com-

bustion could be maintained within 10 minutes without the need for ignition and, in half an hour, operators were assured of continued combustion from the hot refractory. The new ring burner design with reduced mixing and with the injection of air close to the refractory wall was the correct answer to the earlier burner problems.

The reactor heat-up with hydrogen for gasification test 6 proceeded very smoothly and more rapidly than with natural gas and air because there was no need to go through high-pressure purges after the reactor achieved process temperature. Test 6 was started on June 2, 1972, with the feeding of lignite slurry into the gasification reactor at 500 psig. The test was terminated June 16 at a reactor pressure of 800 psig. More than 60 tons of lignite were fed and an average methane concentration of approximately 20% on a dry, nitrogen-free basis was obtained during reasonably steady periods.

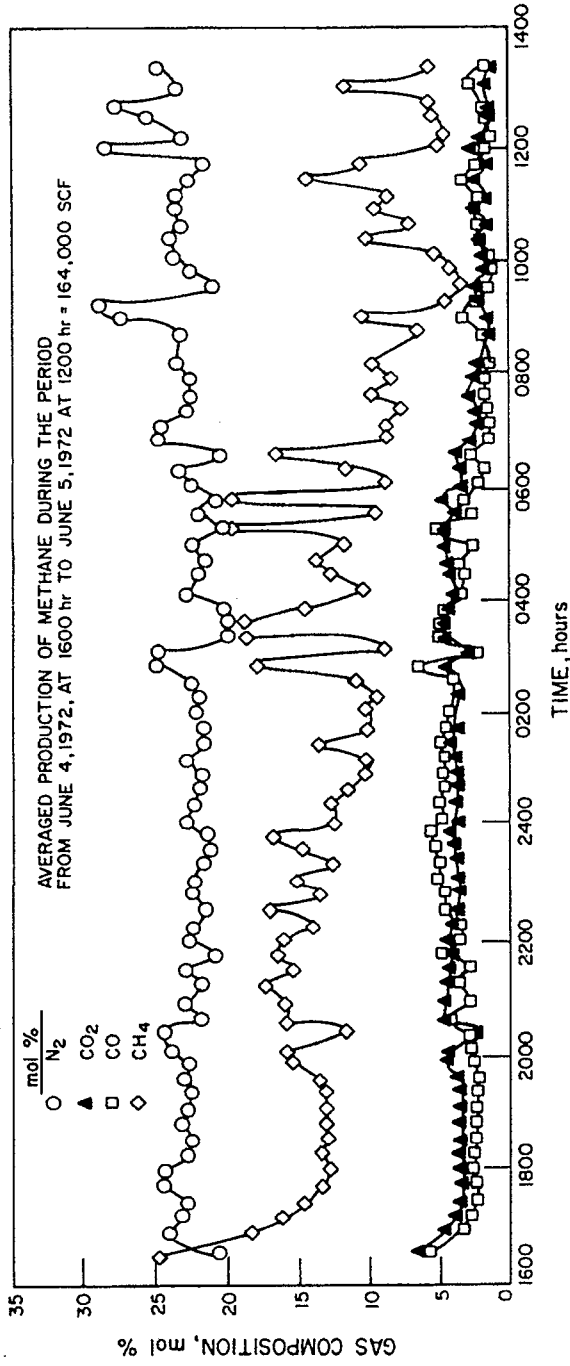
On June 16, the reactor pressure was raised to 800 psig. The temperature in the second-stage gasifier rose significantly, indicating the onset of the exothermic carbon-hydrogen reaction to form methane. At the lower pressures, such a rise was not noticeable because of the lower hydrogen partial pressure. The presence of methane formation reaction with the more graphitic carbon was very encouraging.

Figure 3-23 shows the composition of dry, quenched effluent gas from the hydrogasifier during the steady-stage period of approximately 20 hours. During this period the coal feed averaged 1 ton per hour. Reactor pressure was 500 psig, and air-hydrogen combustion was used to maintain a temperature ranging from 800° to 1200°F in the first-stage lift line, and from 1300° to 1600°F in the fluidized-bed second stage. The methane make averaged 8000 standard cubic feet per ton of coal fed.

Figure 3-24 shows operation at 800 psig on June 16. The exothermic reaction occurred from 1600 to 1900 hours. The significant rise in the methane concentration clearly indicates second-phase or slow-rate carbon gasification.

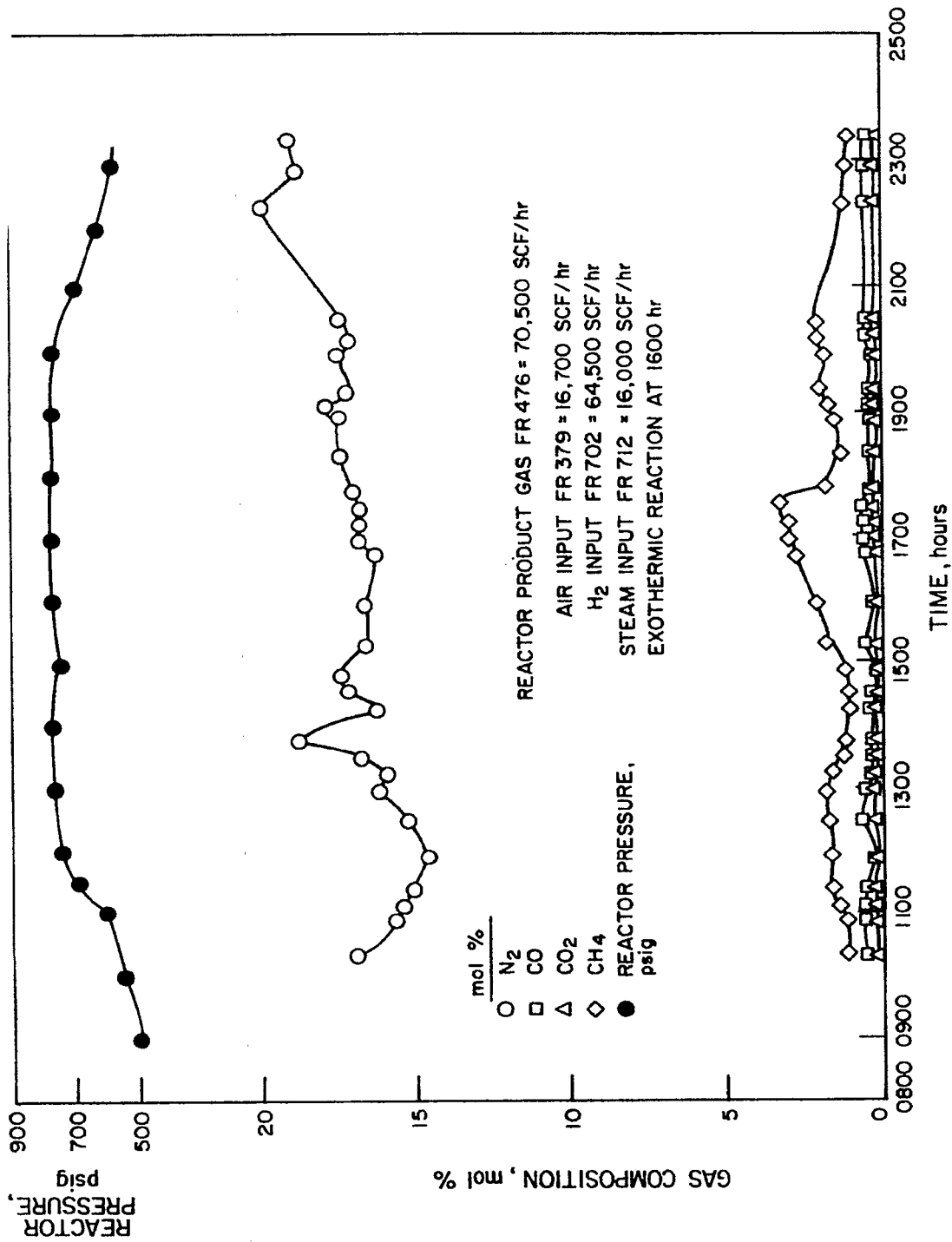
Solids were moved all the way through the reactor and out through the slurry discharge without any plugging for a significant period of time. After this period, however, problems with solids flow from stage to stage interrupted the slurry feed. The first lag in flow occurred in solids movement from the heat exchange bed out to the discharge system. After several days of operation, engineers concluded that the heat-exchange bed around the discharge valve was empty and that the pressure drop indicated in the bed reflected some buildup, constriction, or agglomerates in the heat-exchange bed.

Nearly two days was spent reestablishing solids flow from the slurry dryer into the lift line. Repeated blasting with high-pressure nitrogen at many available locations finally moved the solids down to the bottom. Once started, however, solids flow could be turned on or off rather easily. Possibly, build-up or agglomerates had formed in the transfer pipe or in some part of the slurry dryer bed. With time, these blockages were broken and the flow could be reestablished. No difficulty was encountered in moving solids from the top of the lift line into the second-stage gasifier during the entire run.



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Figure 3-23 GAS COMPOSITION DURING STEADY-STATE PERIOD IN GASIFICATION TEST 6



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Figure 3-24. GAS COMPOSITION AND REACTOR PRESSURE VERSUS TIME DURING JUNE 16, 1972, GASIFICATION TEST 6

Solids flow could not be reestablished from the second-stage gasifier into the heat-exchange bed. Again, flow through this line was smooth and easy to establish in the beginning. The problem appeared to be in the transfer pipe, because no gas bypassing was noticed around the grid.

Because of the solids-flow problem and that of the hydrogen compressor, the test could not be continued on a steady basis. The operators therefore went to 850 psig, the maximum pressure the compressor could provide, and operated the gasifier at 800 psig to get some feel for the behavior of the equipment at pressures higher than had been utilized in operations to that date. Operators also wanted to get some indication of the gasification reaction at a higher pressure.

The slurry dryer operated well during some periods, but, at other times, showed unresponsive bed-level control. Except for one period during which a slug of solids carried over into the quench system, the fines carry-over, which plagued the last test, was not evident. The quench separation, using a new nonionic surfactant additive, was good. Interface control could be maintained at all times except for the one period mentioned above. Even after this surge of fines, however, operators were able to clear the system of coal by purging with water, and the plant returned to stable operation.

The hydrogasifier vessel was cooled down without any signs of rupturing pipes which had accompanied the cooldown periods of earlier tests. The reactor was opened and cleaned out. The slurry dryer bed appeared to be in good condition; however, the extended slurry inlet pipe worked its way loose from its threads and fell to the bottom of the slurry dryer. After that, the slurry entered the bed through an open pipe and may have contributed to difficulty in detecting the proper bed level. In addition, the pressure-drop measurement across the bed and the radiation-level gage both showed a residual reading on an apparently empty bed when the reactor was cold. During the final nitrogen purge, however, and prior to opening the reactor, both indicators had dropped back to zero, leaving engineers without a clue concerning the reason for the high residual reading.

The first- and second-stage gasifiers were both clean, no clinker formations were present. The heat-exchange bed contained large amounts of melted metal which came from the ring burner and the section of the transfer pipe from the second stage. Looking at the history of this test, there were two periods when hydrogen flow was diverted unintentionally from the heat-exchange bed. During these periods engineers believed, the combustion of air through the ring burner was allowed to approach stoichiometric ratios. The resultant high temperature melted the burner-ring support, the ring, and the transfer pipe. This was believed to be the explanation for an inability, at the end of the test, to move solids from the second-stage gasifier into the heat-exchange bed. The molten metal, slagged with lignite ash, formed a canopy across the reactor which showed up as a positive pressure drop. At the time, this pressure drop was interpreted as the presence of a bed in the heat-exchange stage. After continued unsuccessful efforts to move the bed, engineers had concluded that the bed was empty and that the pressure drop was caused by some blockage, such as the one that was found.

The Wilfley slurry-circulating pump was reassembled after a home-made gasket was cut to replace the rubber gasket that had come with the pump. The pump operated well, except that it delivered less flow than the earlier Ingersoll-Rand pump at the same back pressure. As a result, a number of plugs occurred in both the suction and discharge lines. Because there was no device in the slurry system to measure the flow directly, operators had to rely on the pump curve to estimate the flow. The choke on the circulating loop was opened to reduce back pressure, but with a larger flow, and no more of the plugging problems were encountered, as in the early part of the test. A magnetic flowmeter was planned for the slurry line to provide an indication of flow rate.

The Wilson-Snyder mud pump with the stainless-steel valve and seats was used for the start of the test. This was taken off the line after two of the three cylinders gradually failed to pump. The spare pump with the Viton valves was then used for almost a full week before the valves became worn and were replaced. Following valve replacement in the spare pump, no particular problem was noted.

The high-pressure slurry pump was opened and examined after the test; the latest Garlock coating of tungsten carbide on an oxidation-resistant substrate had chipped and peeled off.

The hydrogen compressor was run at 900 psig. High discharge temperatures and noise in the third stage caused engineers to have it disassembled before this test. The compression rings were found to be severely worn. The worst mean spots were 0.287 inch thick compared to a starting thickness of 0.490 inch. These rings could not be reused. The packing rings on the third stage were also badly worn; hydrogen leaks through the packing vents were detected. The compressor was reassembled with a new set of packing rings and a new set of compression rings of the old diagonal-cut design, because there were no more rings of the new custom-machined overlapped stepped-joint design. In the meantime, an order was placed with Ingersoll-Rand to supply all of the necessary equipment to convert this machine to a minilubricated unit. The lubricators, tubing, and drives, as well as instructions for assembly, were shipped by June 16. An oil droplet separator was also ordered, to be installed at the discharge of the third stage to remove as much of the oil added as possible. A high-temperature lubricant (Tenneco Anderol 500) was then selected for use to further reduce the amount of oily vapor which may enter the downstream hydrogen superheater. Plans were made to adjust the firing temperature on the superheater to avoid, as much as possible, the cracking of the lubricant vapor.

The problem with the valves was analyzed and Ingersoll-Rand supplied the HYGAS project with plate valves instead of channel valves, for the first and second stages. The plate valves on the third stage had not caused any problem to that date. It seemed reasonable, then, to try the same design for the first and second stages. The rider rings on all the stages were in good condition, as were the piston rods and the cylinder walls.

The hydrogen compressor was reassembled with valves, valve adapters, and new compression rings. The machine was started on May 19 and was

run at 900 psig through the weekend. The hydrogen compressor was disassembled for inspection on May 22, found to be in good condition, and was reassembled. Upon continued running between pressure ranges of 900 and 1300 psig, excessive noise was detected in the third stage on May 26. An inspection showed that the compression rings had been worn as much as 0.050 inch. The allowable wear on these rings was approximately 0.250 inch. The excessive wear was obviously unacceptable, and Ingersoll-Rand was contacted to determine what the next step should be.

The coal crushing equipment produced enough coal to continue operations. Operation improved, but was not perfect. Williams advised HYGAS engineers that the wind box, which was bolted to the mill and was leaking in large amounts of air because of the buckling plates, should have been welded to the mill; this was not an easy task because of access problems and dissimilar metals. During the test, operation continued with the leaky areas patched with high-temperature sealant. From 14 to 16% of -100 mesh coal was being produced and was quite satisfactory for start-up operations; however, the 15 to 18% of moisture content in the coal was not as low as desired.

After test No. 5, the wind box was welded to the mill on the Williams coal mill; the same thing had been done at the Rapid City plant. It was believed that the welding would eliminate air leakage into the mill and would help provide a proper ΔP reading in the coal mill during operation. The coal mill was run once after the wind box had been welded; performance was satisfactory, except that the fines generation was high. The velocity separator was adjusted to obtain a better size distribution.

During test No. 5, work was performed on the quench separation clean-up, and tall oil was added to the circulating water to break up any emulsion that might be formed. An entire series of surfactants and additives was investigated at this time, in a search for means to improve the settling characteristics of solids from either the oil or the water phase. The objective was to prevent the collecting of a layer of solids between the water and oil phase that would directly affect the operation of the interface controller.

The discharge char-water slurry filter was activated. Mechanically, the filter operated satisfactorily; however, a steady concentration of solids was not provided as feed to the filter. Engineers were convinced that, once the char-water slurry discharge stabilized, the filter would operate correctly.

For Test No. 6, the hydrogen compressor was operated at as low a pressure as possible to feed only necessary hydrogen to the reactor; the reduced operation was to prolong the life of the rings. The packing on the third stage leaked, indicating wear. In addition, the second- and third-stage discharge temperatures were higher than the theoretical temperature, indicating gas bypass across the piston rings. On the other hand, the first-stage discharge temperature coincided almost exactly with the theoretical temperature. Canal water was used to cool the third-stage cylinder to maintain less than 10°F rise across the cylinder, as specified by Ingersoll-Rand. Because operators could not get a pressure greater than 850 psig - the discharge pressure near the end of the test - this certainly indicated substantial wear on the piston rings, for the third stage.

After the test, the hydrogen compressor was disassembled. The packing rings were completely worn through on the third stage, and the piston rings on the third stage were severely worn and cracked.

3.5.11 Annual Plant Turnaround, July, 1972

The plant was shut down for two weeks during July for plant turnaround. Many items of work related to maintenance and modifications were accomplished, particularly those involving utility tie-ins which could not be made until the boiler was down.

The hydrogasifier vessel was cleaned out after Test 6. The baffle for the gas-discharge line, which was not put in before the last test because it was not ready, was installed in the slurry dryer. Two additional pressure taps were installed in the slurry dryer bed to measure bed density and level.

The blast connections on the transfer valves between the slurry dryer bed and the lift line, and between the lift line and second stage, were moved up into the pipe to provide a more effective blast. The location of the lowest tap was believed to have caused the difficulty in moving solids through the transfer line during the test.

The ring burner in the heat-exchange bed was rebuilt. An additional purge directly above the transfer valve from the heat-exchange bed was installed to facilitate solids removal.

The 18-inch-diameter spent-char slurry quench drum was enlarged with a 36-inch-diameter lower section. This increased the slurry inventory so that, when solids discharged from the hydrogasifier during a start-up, the concentration change in the slurry inventory would be less abrupt than in earlier tests. The enlarged vessel was also designed to eliminate problems of solids plugging in the discharge slurry lines. A weld X-ray inspection of this section proved satisfactory. A hydro-test was performed on the vessel to 1.5 times the working pressure, and the final piping tie-ins were completed.

The conversion of the HHE hydrogen compressor to "minilube" operation was completed under the supervision of an Ingersoll-Rand field engineer. A new heat exchanger and oil mist separator, and associated piping, were also installed. The HHE compressor was started up under "minilube" operation and was tested up to its rated working pressure of 1500 psig. Under the break-in operating schedule, operation began at a gradually increasing discharge pressure until the 1500-psig discharge was reached.

The slurry system modifications were complete. A magnetic flowmeter had been installed in the low-pressure slurry loop to try to determine the flow, even though pure toluene might not have sufficient electrical conductivity for a magnetic flowmeter. The piping modifications on the high-pressure slurry system were completed to provide smoother and safer switching of slurry in and out of the reactor. The high-pressure slurry circulating pump was sent out for Stellite coating of the casing and the base metal bond with stellite was an improvement over the flame-sprayed coatings, which appeared to have been inadequately bonded to the base.

The pretreatment section was tested during the week of July 3 with Illinois No. 6 bituminous coal. The new venturi scrubber operated well, but difficulties occurred with the new solids-feed device — a heavy-duty Gemco valve operating on a lock-hopper basis. The valve shaft, operating through nonlubricated sleeve bearings, seized and galled; after numerous attempts at substitution repairs, the eccentric bushings that supported the shaft had to be modified. Teflon parts were used for the modification instead of cast iron. The test with Illinois No. 6 coal produced nonagglomerating product.

The major revisions in the low-pressure steam supply and condensate return systems ensured the better operation of the HYGAS steam-tracing system and better plant "winterizing".

3.5.12 Gasification Test No. 7, August, 1972

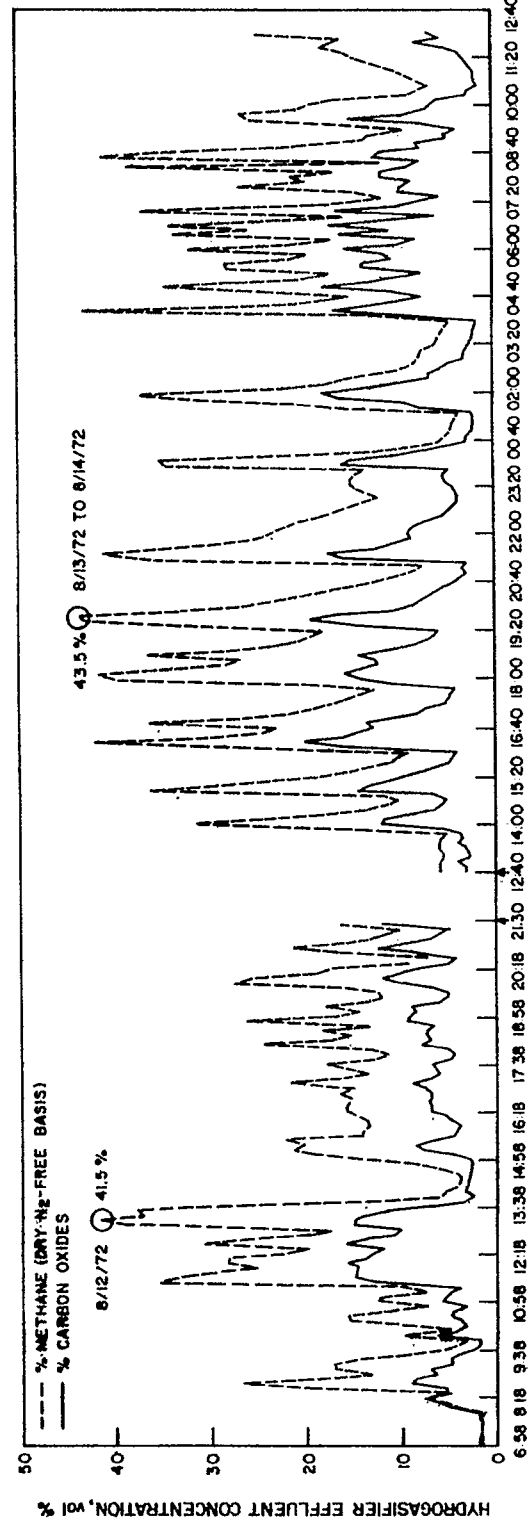
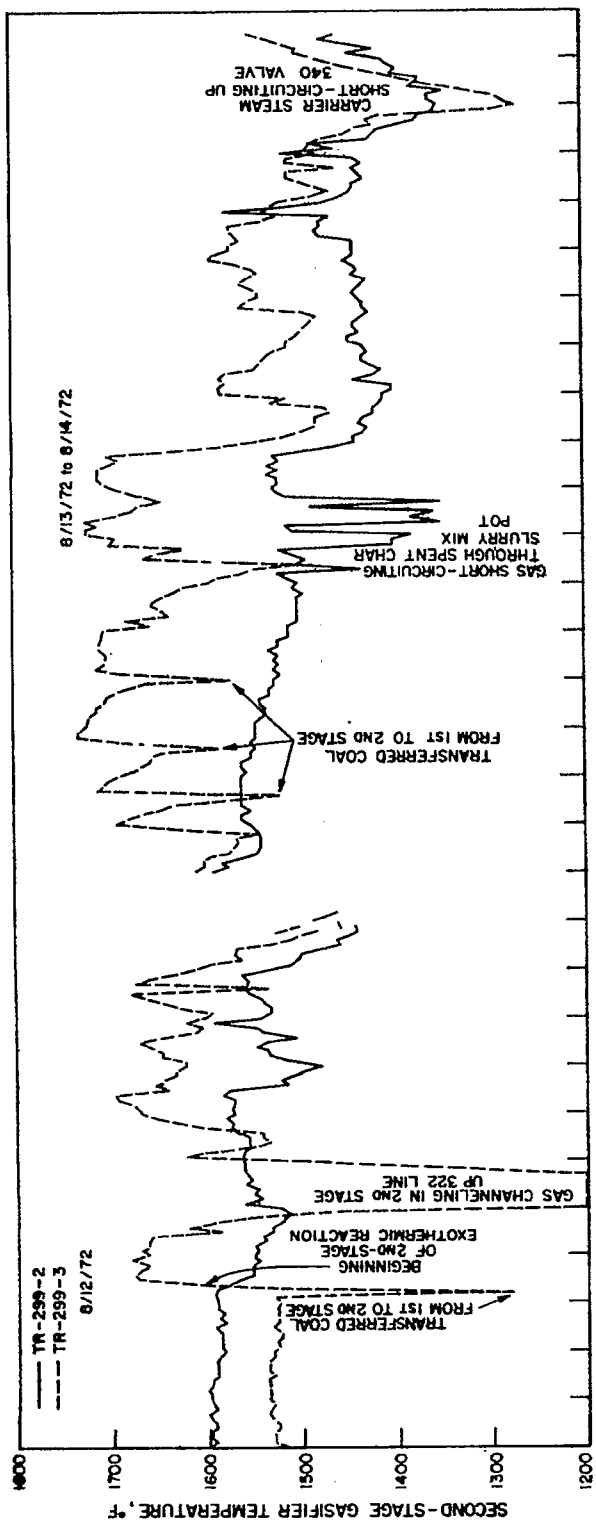
The gasifier was operated at 1000 psig for the first time. This is the pressure at which balanced gasification reactions take place, and the pressure upon which commercial plant designs are based. Overall, the equipment performed well at the highest pressure yet reached in start-up testing.

At 1000 psig, sufficiently high hydrogen partial-pressure was achieved for the second-stage gasification to occur. This methane formation reaction is exothermic, and there were positive indications that it had begun in the form of temperature rises of as much as 150°F in the second stage. Temperatures of 1750°F or slightly higher were achieved. For the very reactive lignite, temperatures of 1600°F should be more than adequate to carry out both the methane formation reaction and the steam-carbon reaction.

With the significant reaction taking place in the first and second stages, the methane concentration in the make gas rose correspondingly. A concentration of more than 40% methane was reached on a dry, nitrogen-free basis. This was just about the design methane concentration from the gasifier. After gas purification, this gas composition would represent the design composition as a feed to the methanator.

Figure 3-25 shows the temperatures in the second-stage gasifier and the methane and carbon oxides concentrations in the gasifier effluent during the two periods of "best" coal feeding during Gasification Test No. 7. Because of the transfer valve problem, coal feed to the second stage was not continuous, resulting in the peaks in temperatures and gas compositions. Table 3-10 presents some data on the gas production rate during these two periods.

After a break-in period, the hydrogen compressor was operated for 48 hours at a 1500-psig discharge pressure. The compressor performed very well during this test with no apparent change in performance at a discharge pressure of 1300 psig over a two-week period. Lubrication of the compressor apparently had solved the compressor problems that had been evident in earlier testing.



D-92-937

Figure 3-25 SECOND-STAGE GASIFIER TEMPERATURE AND CONCENTRATIONS OF METHANE AND CARBON OXIDES IN THE GASIFIER EFFLUENT DURING THE BEST COAL-FEEDING PERIODS

Table 3-10. GASIFICATION TEST No. 7

| | Period 1 † 8/12/72 (14.5 hr) | Period 2 ‡ 8/13 to 8/14 (23.6 hr) |
|---|------------------------------------|---|
| Total Gas Effluent Flow, SCF | 1.02 X 10 ⁶ | 1.69 X 10 ⁶ |
| Avg Gas Effluent Flow, SCF/hr | 7.02 X 10 ⁴ | 7.15 X 10 ⁴ |
| Total CH ₄ Production, SCF | 1.14 X 10 ⁵ | 2.31 X 10 ⁵ |
| Avg CH ₄ Production, SCF/hr | 7.81 X 10 ³ | 9.80 X 10 ³ |
| Total Carbon Oxides Production, SCF/hr | 5.06 X 10 ⁴ | 9.82 X 10 ⁴ |
| Avg Carbon Oxides Production, SCF/hr | 3.48 X 10 ³ | 4.16 X 10 ³ |
| Total Carbon Gasified, lb | 5300 | 10,600 |
| Avg Carbon Gasified, lb/hr | 367 | 450 |
| Total Coal Fed, lb | 15,500 | 43,600 |
| Avg Coal Feed Rate, lb/hr | 1070 | 1850 |
| Carbon Gasification,* % | 61 | 43 |
| Methane Produced/Coal Fed, SCF/lb | 7.33 | 5.31 |
| Carbon Oxides Produced/Coal Fed, SCF/lb | 3.27 | 2.25 |

* Based on Run 5 feed analysis

† Period 1: 6:58 a.m. 8/12/72 to 21:30 p.m. 8/12/72.

‡ Period 2: 12:40 p.m. 8/13/72 to 12:04 p.m. 8/14/72.

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The gas purification section was activated for the first time by passing process gas through it. Although an apparent partial blockage in the absorber caused liquid carryover with the off-gas, all the other equipment appeared to be in good operating condition at 1000 psig pressure.

The new slurry drum, with the enlarged bottom section, smoothed out the surges in slurry concentration. Solids discharge from the bottom of the hydrogasifier, with the addition of a new aeration tap above LV 340, simplified the initiation of flow.

The coal mill operated well. Changing the flame detector location on the furnace has eliminated the periodic erroneous shutdowns experienced earlier due to faulty flameout detection.

After the test, the various sections of the reactor were examined and improvements were begun in areas that had given trouble during the tests. The results of the inspection were:

- 1) The hydrogasifier vessel was opened up and cleaned. A consultant from Bell Aerospace reviewed the hydraulic system used to control solids transfer valves. A number of good suggestions were made to ensure operability and to simplify the equipment. These were incorporated before the next test. Otherwise, the reactor interior looked good. The solids withdrawal area near the bottom of the heat-exchange bed was modified to improve solids discharge. A standpipe was added to increase the seal leg above the solids-discharge valve, and to eliminate the turbulent area near the hydrogen-steam inlet ring.
- 2) The packing in the bottom section of the absorber had melted when the quench water to the off-gas quench tower had to be stopped for a short period to isolate a badly leaking flange gasket. The polypropylene packings were removed very slowly by drilling and jack-hammering. The packing was replaced with stainless steel. Because the quench water stoppage occurred after an initial attempt at putting the purification system on-stream, the cause for the main carry-over from the absorber was not yet determined. Engineers suspected that the bottom distributor tray had been partially blocked.
- 3) The hydrogen compressor was disassembled and thoroughly inspected. Measurements indicated there was no wear, and the unit was reassembled, ready for the next operation.
- 4) The light oil-water quench separator was partially filled with a mixture of water and coal fines. The amount of coal fines was not large in terms of the total coal fed to the gasifier; however, this mixture occupied enough of the separator volume that good separation had not been achieved near the end of the test. Study was begun of a number of possible solutions.
- 5) The high-pressure discharge slurry pump was inspected. The Stellite inlay put on the pump held up extremely well, showing no sign of any wear. Although the test period had not been long enough to establish long-term effects, the new inlay was much more promising than any of the coatings previously used on this pump.

APPENDIX 3-A
Materials and Corrosion
Considerations in the
HYGAS Process

3-Ai

Editor's Note

This discussion, presented before a National Science Foundation Workshop, is presented here to provide additional insight into hydrogasification materials problems.

MATERIALS AND CORROSION
CONSIDERATIONS IN THE
IGT HYGAS PROCESS

by

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Prepared for Presentation to
A National Science Foundation Workshop
on
Materials Problems and Research Opportunities
in Coal Gasification

at

The Ohio State University
Columbus, Ohio

April 16-18, 1974

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SUMMARY

The HYGAS Process for conversion of coal to high-Btu pipeline-quality gas has reached an advanced stage of development during two years of pilot testing at the IGT pilot plant in Chicago. Clean SNG has been produced in quantity during sustained operation of all plant units, with heat values on occasion exceeding 1000 Btu/SCF.

Coal of any rank or type may be utilized as HYGAS plant feed. Temperature and pressure in the continuous two-stage process are 1200° to 1400°F in the first reactor stage at 1100 psig, and 1600° to 1800°F in the second reactor stage at 1100 psig.

Three processes are under investigation by IGT at the same pilot facility to provide hydrogen-rich gases for use in the HYGAS Process. The emphasis over the next year or two will be on the utilization of the steam-oxygen system; this system is used as a point of discussion. Characteristics of the HYGAS Process with these hydrogen-producing processes are:

HYGAS and Steam-Oxygen Gasification of Char: Heat source is the oxygen-char heat-of-reaction; utilization of an oxygen plant provides a nitrogen barrier; the gas-solids contact modes are suspension and fluidized bed; the technique for ash removal — to assure high carbon utilization — is an agglomerated-ash bottom.

HYGAS and Steam-Iron Gasification of Char: Heat source is the air and char reaction in a producer vessel; iron and iron-oxide provide the nitrogen barrier; contact modes and ash removal are the same as with the steam-oxygen process.

HYGAS and Electrothermal Gasification of Char: Heat source is electrical energy; the nitrogen barrier is electricity; high carbon utilization is guaranteed through the heat value of by-product char, subsequently used for fuel; the contact modes are the same as with the steam-oxygen process.

The presence of steam and hydrogen at high pressures and temperatures in this process (as in virtually all coal gasification processes) creates challenges in the prevention of corrosion, including those problems that result from hydrogen embrittlement. Extensive data concerning corrosion have been collected by IGT in its two years of pilot plant operations, and typical corrosion data are presented.

THE HYGAS PROCESS

The HYGAS Process (Figure 1), developed by the Institute of Gas Technology (Chicago) for the U. S. Department of the Interior's Office of Coal Research, and the American Gas Association, has reached the pilot plant stage. The pilot plant is designed to process 75 tons of coal and produce 1.5 million cubic feet of pipeline-quality gas daily. The process can operate with any rank or type of coal. The process is tailored for maximum thermal efficiency by maximizing the direct formation of methane in the reactor. High temperatures (1200° to 1400°F in the first stage, and 1600° to 1800°F in the second stage) guarantee reasonable reaction rates, and high pressure (about 1100 psi in both stages) increases equilibrium methane yield.

The most active fraction of coal is hydrogasified to form methane, while the less active fraction is used to generate hydrogen in support of the reactions. The raw gas from the hydrogasifier contains substantial amounts of carbon monoxide and hydrogen. These are converted indirectly to methane during catalytic clean-up methanation, boosting the heating value of the gas, and reducing carbon monoxide content to below 0.1%.

Basic process steps include:

- Coal receiving and storage.
- Coal processing into the size required for gasification, and into a coal-oil slurry for convenient introduction into the coal gasification reactor.
- Coal gasification, where gas is generated at high temperature and pressure. The principal reactions of interest within the reactor are:



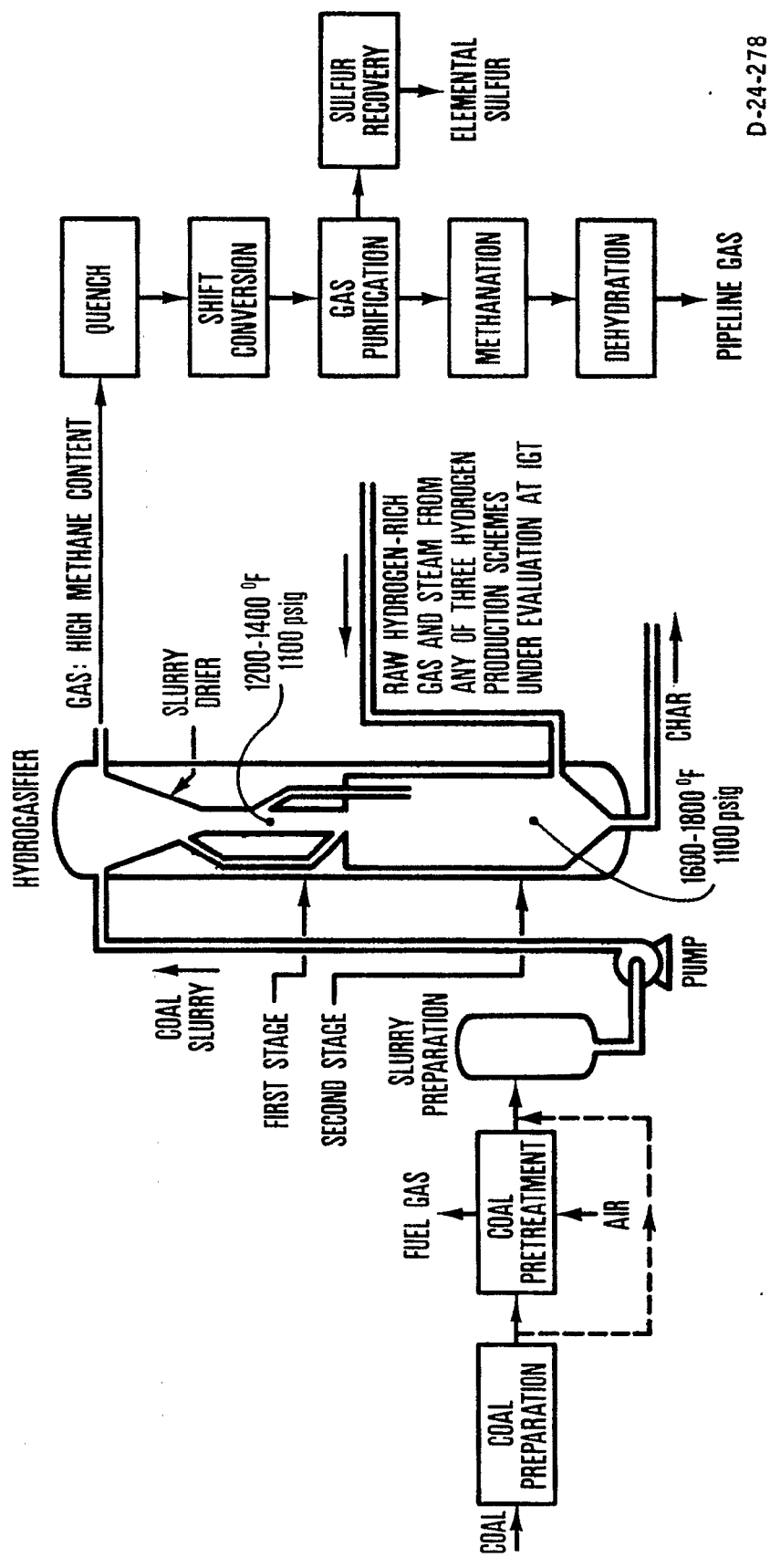
which forms methane directly, and:



which forms the carbon monoxide and hydrogen (synthesis gas) which is later catalytically converted to additional methane.

Continuing the list of basic steps are:

- Gas scrubbing and cooling.



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Figure 1. SIMPLIFIED REACTOR CUTAWAY AND BLOCK DIAGRAM OF THE IGT HYGAS PROCESS

- Shift conversion of carbon monoxide in the gas to carbon dioxide, and the generation of more hydrogen.
- Acid-gas removal and purification, taking out carbon dioxide and hydrogen sulfide.
- Methanation, to maximize the heating value of the gas by reacting hydrogen and carbon monoxide to form additional methane:



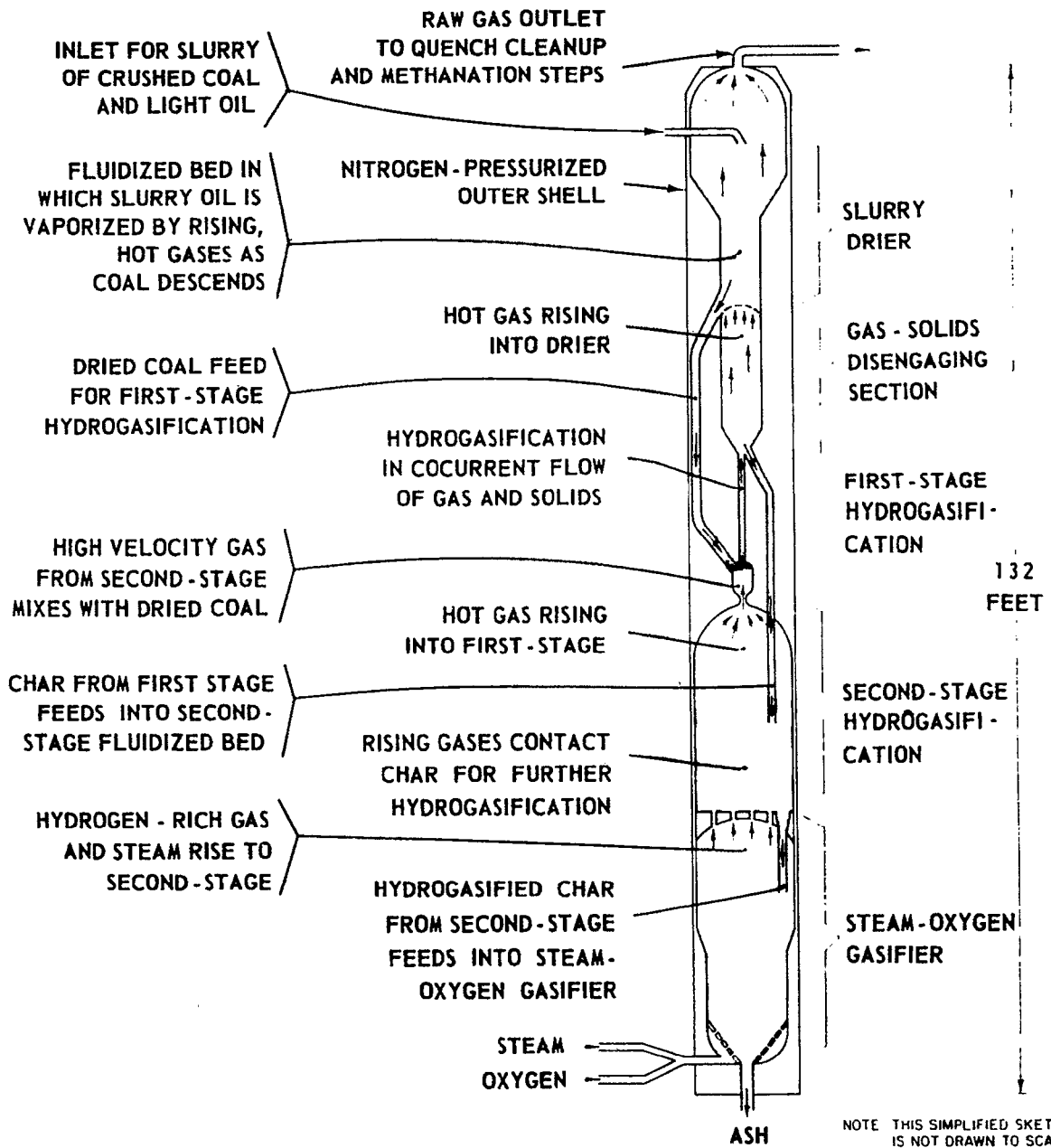
- Dehydration to meet pipeline specifications for a clean gas with high-Btu/SCF value; the gas is already at pipeline pressure.

PROCESS STEPS

In the HYGAS pilot plant operations, coal is ground to -8 mesh size and fed into a pretreater if the feed is an agglomerating coal; it is fed directly to the slurry preparation section if coal is non-agglomerating. The caking tendency of coal is destroyed in the pretreater by mild surface oxidation of the coal particles, using air as the oxidant; a free-flowing coal results. All coal, with or without pretreatment, is slurried with a by-product aromatic oil and then pumped to a pressure of 1200 psi and moved to the reactor (Figure 2). The oil is evaporated in a slurry drier, and later condensed and recycled. The dried coal is moved to the bottom of the first hydrogasification stage.

In the first, low-temperature reactor stage, coal enters a high-velocity rising stream of hot gases that originate in the lower, second stage. As the solids/gas mixture rises, the coal is rapidly heated by dilute-phase contact with hot reaction gases. At the top of this riser-reactor, gas velocity is reduced, solids drop out, and gas continues upward, drying incoming coal in the slurry drier. Hot char is channeled downward to a dense-phase fluidized bed in the second, high-temperature reactor stage. Hot gases pass through the fluidized bed contacting char with good mixing. Organic carbon remaining in char after this stage can be used at other process points to produce both hydrogen and heat required in the HYGAS conversion process. Subsequent process steps include quench, shift to a 3:1 ratio of hydrogen to carbon monoxide, gas purification, methanation, and drying before the final product emerges as high-Btu SNG.

A simplified process flow sheet is presented in Figure 3.



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Figure 2. SIMPLIFIED SKETCH OF IGT PILOT PLANT HYDROGASIFICATION REACTOR SECTION

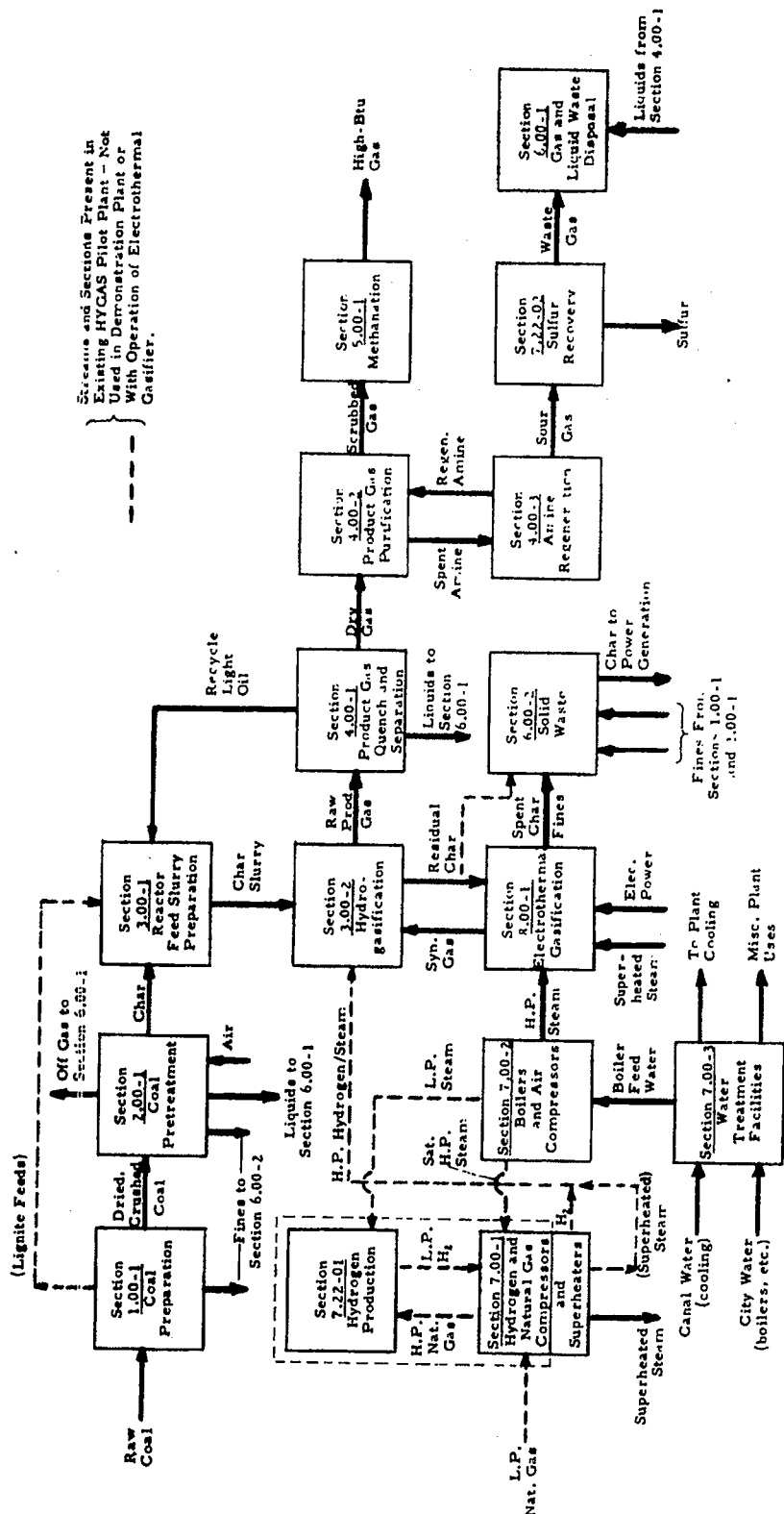


Figure 3. HYGAS PILOT PLANT PROCESS FLOWSHEET

HYDROGEN-RICH GASES

The production of hydrogen-rich gases needed to produce methane in the HYGAS and other processes is of major technical and economic importance. Three means to produce hydrogen-rich gases are being investigated at the IGT pilot plant. The hydrogen-rich gas producing processes are Steam-Oxygen Char Gasification (Figure 4A), Steam-Iron Char Gasification (Figure 4B), and Electrothermal Char-Gasification (Figure 4C). Emphasis during the next year or two will be on utilization of the steam-oxygen process; this system is used as a point of discussion.

Steam-Oxygen Gasification

Excess char from the HYGAS reactor can be reacted with steam and oxygen in a high-pressure fluidized bed. The quantities of steam and oxygen used relative to the char are controlled so as to establish an operating temperature of about 1800° to 1900°F. The reactor, now being assembled at the HYGAS pilot plant, will be a simple, internally insulated vessel.

The novel feature of this development is the gasification of char under nonslagging conditions in a high-pressure fluidized-bed which, at the same time, achieves high carbon utilization and significant methane formation, with low oxygen consumption. The new steam-oxygen gasifier now under development will be integrated into HYGAS pilot plant operations during 1974.

A HYGAS Process flowsheet incorporating steam-oxygen gasification is presented in Figure 5.

Steam-Iron Gasification

Historically, an early form of the steam-iron process to produce hydrogen was operated at atmospheric pressure using two beds of iron solids. One bed was reduced from iron oxide to iron by a suitable gas; the second bed was simultaneously oxidized by steam from iron to iron oxide. Hydrogen is produced in this steam oxidation of iron. The new IGT process replaces the historical, cyclic operation, which used two static beds, by using fluid-bed reactors in which iron material continuously circulates between a reductor and an oxidizer. The new process operates at HYGAS pressure so that the virtually pure high-temperature hydrogen-steam mixture produced can be passed directly into the HYGAS reactor.

Fig. 4A

STEAM-OXYGEN GASIFICATION

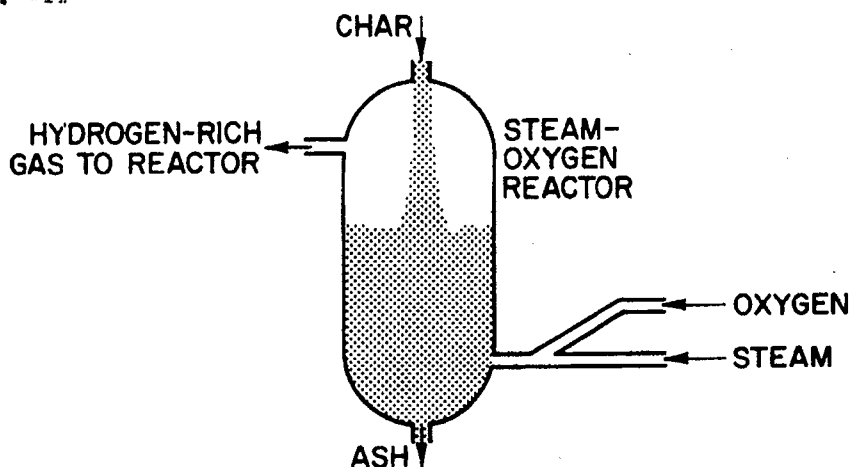


Fig. 4B

STEAM-IRON GASIFICATION

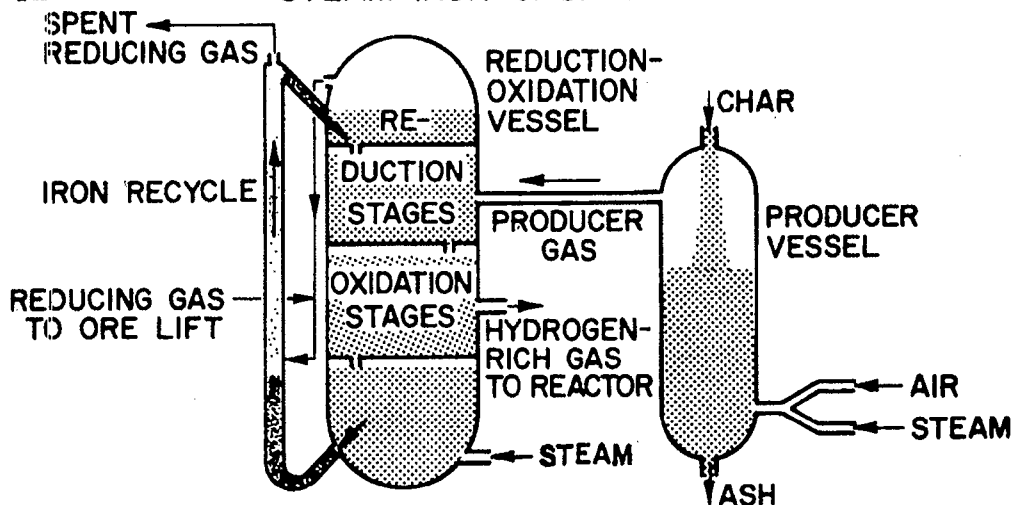
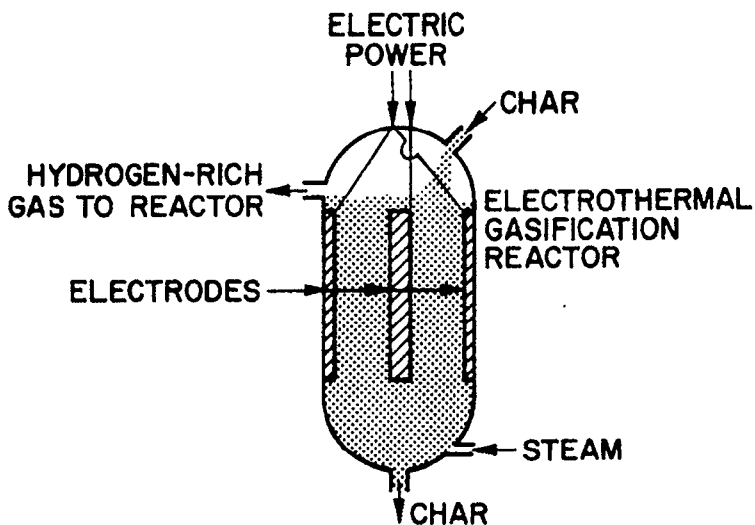


Fig. 4C

ELECTROTHERMAL GASIFICATION



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Figure 4. THREE TECHNIQUES UNDER INVESTIGATION AT THE IGT PILOT PLANT TO PRODUCE HYDROGEN-RICH GAS

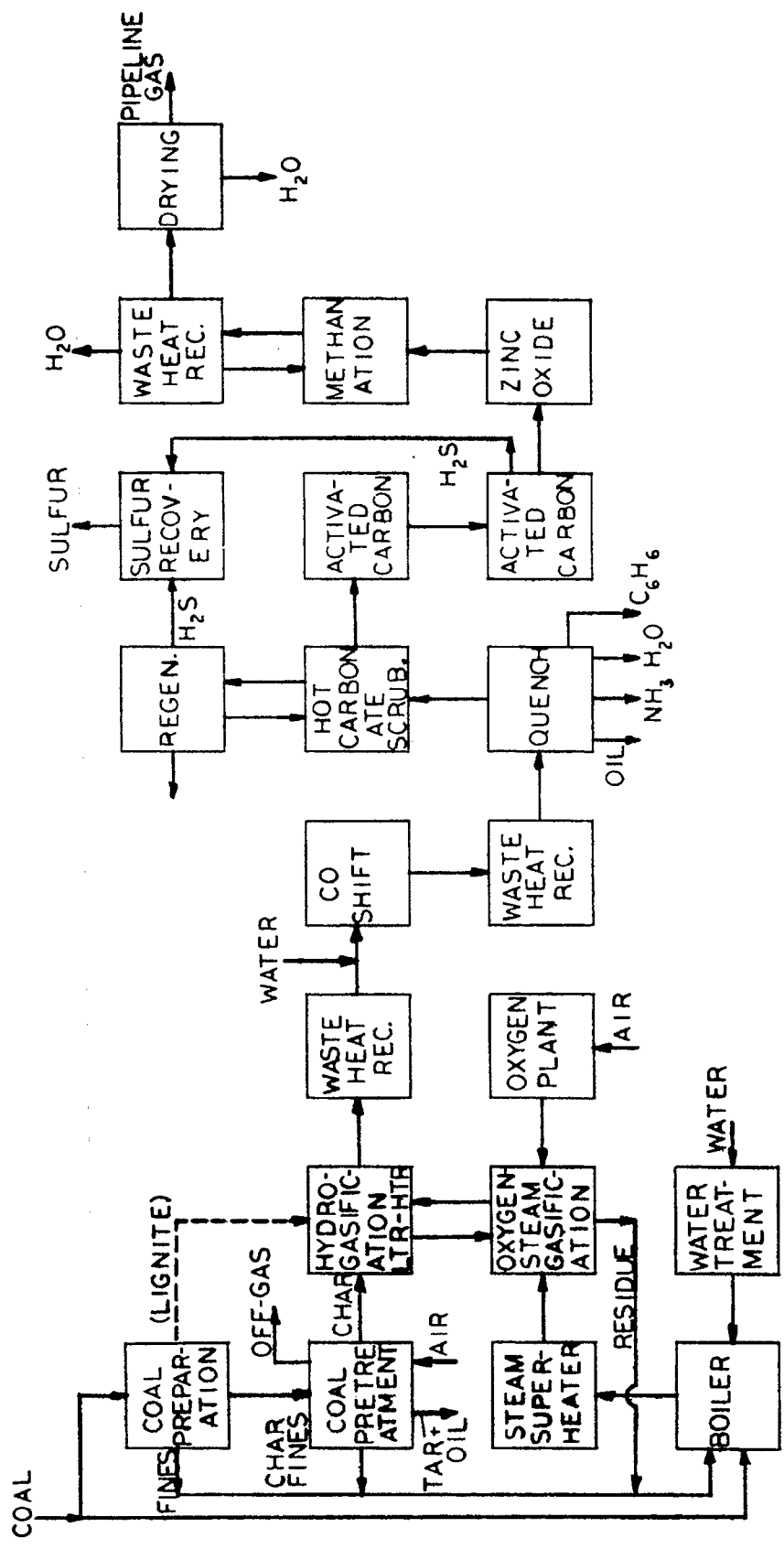


Figure 5. PROCESS FLOWSHEET FOR HYGAS PROCESS COMBINED WITH STEAM-OXYGEN GASIFICATION

Electrothermal Gasification

Char from the HYGAS reactor reacts with steam in a fluidized-bed electrothermal gasifier to form a hydrogen-rich gas which is passed directly into the hydrogasifier. The steam-char reaction is highly endothermic (requires heat); the required heat is furnished by passing direct current through the fluidized bed of char. Residual char from this electrothermal reactor would be used to produce the electric power required, although at the HYGAS pilot plant, the electrical power is purchased. Tests to date at the HYGAS pilot plant indicate that this process is technically feasible and has many attractive features. However, the cost of electrical power generation today, compared with the period when this concept was envisioned, tends to make the two other IGT hydrogen-rich gas production schemes presently more attractive.

MATERIALS AND CORROSION

Designers must guard against possible severe corrosion from a wide range of substances. The principal threat is from hydrogen, steam, and hydrogen sulfide at high temperatures and pressures, at various points in the process. Other potential corrosives and erosives include humic acids, char (corrosive/erosive), aqueous scrubbing solutions and dissolved acid gases, dilute caustics, wet product gas, and slurry (abrasive).

Potential corrosion areas in the HYGAS pilot plant are analyzed in Table 1, by plant stream and section, including pressure range, temperature range, stream composition, corrosive/erosive agents, and materials of construction.

Potential corrosion areas in the HYGAS Process with synthesis gas generated by fluidized-bed oxygen-steam gasification of residual char are presented in detail in Table 2.

Figure 6 is a flowsheet of a hypothetical HYGAS demonstration plant, where potential problems of corrosion and erosion are at least as severe as in the HYGAS pilot plant. The potential corrosion/erosion areas, and agents, together with other pertinent data, are detailed in Table 3.

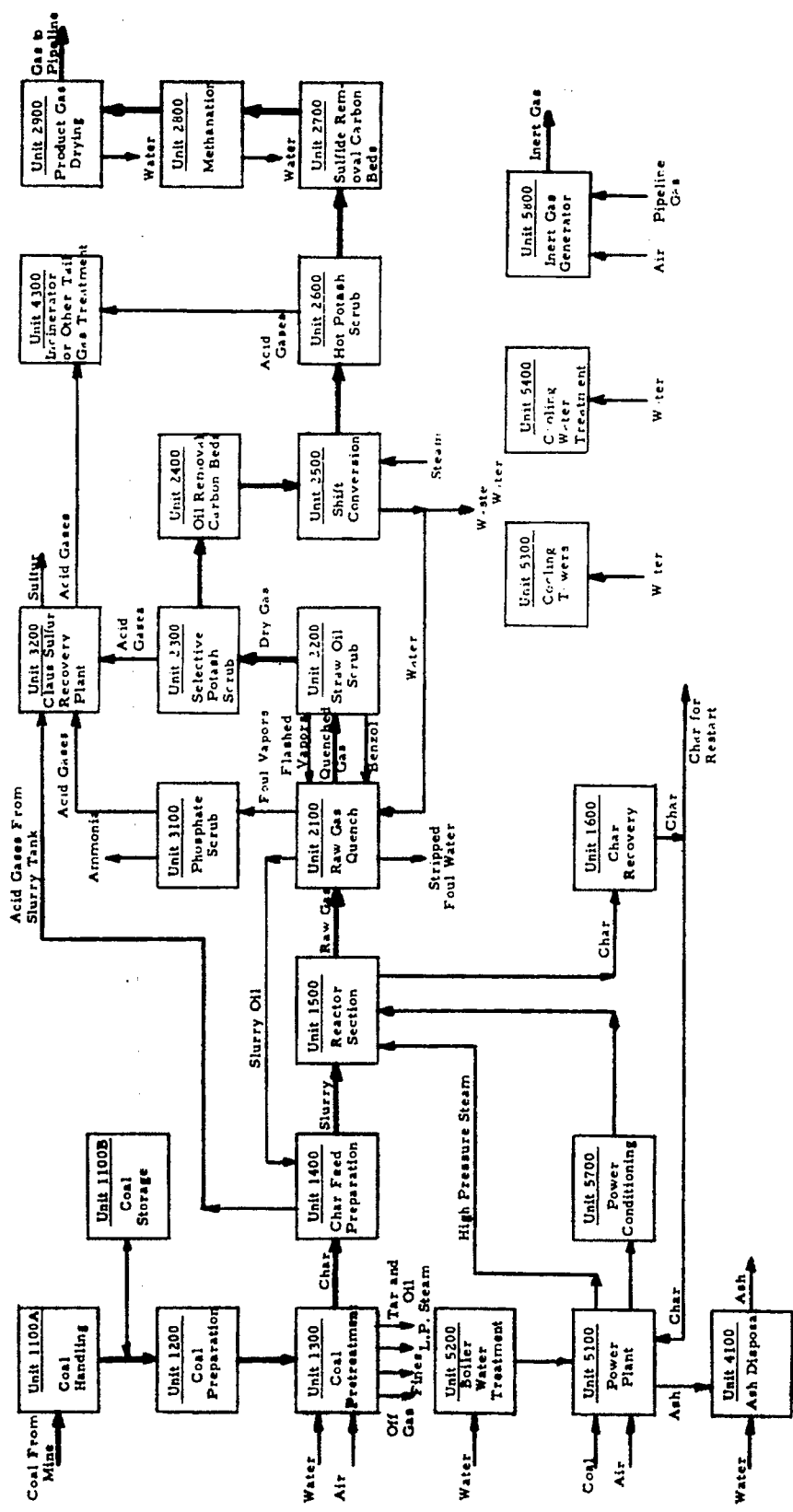


Figure 6. HYGAS DEMONSTRATION PLANT PROCESS FLOWSHEET

Table 1, Part 1. POTENTIAL CORROSION AREAS IN
HYGAS PILOT PLANT

| Stream/Equipment Description | Conditions | | Composition, Mole % or wt % | Material of Construction | Corrosive, Erosive Agents |
|---|--------------------|---------------------------|---|--|---|
| | Temp. Range, °F | Press. Range, P.s.i.g. | | | |
| 1. Section 1.00-1, Coal Preparation | | | | | |
| No special problems are anticipated in this section of the plant and carbon steel (A-285-C for vessels) or equivalent has been specified, except for brand name equipment items or noncustom equipment. However, most equipment must be able to withstand normal atmospheric corrosion since it is not enclosed. Also, to dry wet lignite, it was necessary to use higher mill temperatures and, thus, perhaps different materials may have to be specified. No other problems are anticipated. | | | | | |
| 2. Section 2.00-1, Coal Pretreatment | | | | | |
| a. <u>Stream 201, Air to Pretreater</u> (heated during start-up of pretreater) | 250-1200 | 10 | Air | Type-304 stainless steel (ASTM A312) | High-temperature air |
| b. <u>Stream 204, Pretreater Off-Gas</u> | 800 | 0.8 | N ₂ 67.13 O ₂ 3.00 CO 6.42 CO ₂ 5.00 CH ₄ 0.14 C ₂ H ₆ 0.14 C ₃ H ₈ 0.14 C ₄ H ₁₀ 0.66 H ₂ O 16.65 SO ₂ 0.86 Total 100.00 (plus tars, oils and char fines) | Low alloy steel (ASTM A335-Pl. C-1/2 Mo) | Hot SO ₂ |
| c. <u>Vessel 2.06-1, Pretreater Reactor</u> (Shell refractory lined, but internal cyclones and cooling coils bare same material as shell) | 800 | 0.8-10 | See Streams 201 and 204, above | Carbon steel (ASTM A285C, F. B. Q.) | Hot SO ₂ and char erosion |
| d. <u>Vessel 2.06-42, Pretreater Char Cooler</u> (shell unlined) | 200-800 | 0.8-10 | See Streams 201 and 204, above | Carbon steel (ASTM A285C, F. B. Q.) | Hot SO ₂ and char erosion |
| e. <u>Vessel 2.06-02, Pretreater Quench Tower</u> | 140-800 | 0.8 | See Streams 204 and 210, below | Carbon steel (ASTM A285-C, F. B. Q.) | Moist SO ₂ , dissolved SO ₂ , and humic acids |
| f. <u>Vessel 2.06-41, Pretreater Quench Separator</u> | 175 | 0.8 | See Streams 206 and 210, below | Carbon steel (ASTM A285-C, F. B. Q.) | Moist SO ₂ , dissolved SO ₂ and humic acids |
| g. <u>Stream 205, Quench Tower Bottoms</u> | 175 | 0.8 | H ₂ O and Humic Acids 99.0 Tars and Oil 0.3 Fines 0.7 Total 100.0 | Carbon steel (ASTM A53 Grade A or B) | Dissolved SO ₂ and humic acids* |

Table 1, Part 2. POTENTIAL CORROSION AREAS IN
HYGAS PILOT PLANT

| Stream/Equipment Description | Conditions | | Material of Construction | Corrosive, Erosive Agents | |
|---|-----------------|------------------------------------|--------------------------------------|--|--|
| | Temp. Range, °F | Press. Range, psig | | | |
| h. Stream 206, Heavy Oil, Tar and Char Fines | 175 | 0.8 | Carbon steel (ASTM A53 Grade A or B) | Humic acids | |
| | | | Composition, mole-% | | |
| | | Tars and Oils and Humic Acids 28.5 | | | |
| | | Char Fines 71.5 | | | |
| | | Total 100.0 | | | |
| i. Stream 210, Quench Tower Off-Gas | 150 | 0.7 | Carbon steel (ASTM A53 Grade A or B) | Moist SO ₂ | |
| | | | Composition, mole-% | | |
| | | | N ₂ 61.07 | | |
| | | | O ₂ 3.91 | | |
| | | | CO 5.85 | | |
| | | | CO ₂ 4.55 | | |
| | | | CH ₄ 0.13 | | |
| | | | C ₂ H ₄ 0.13 | | |
| | | | C ₃ H ₈ 0.60 | | |
| | | | H ₂ O 22.97 | | |
| | | SO ₂ 0.78 | | | |
| | | Total 100.00 | | | |
| 3. Section 3.00-1, Reactor Feed Slurry Preparation | | | | | |
| Since the only components present in this section are coal char (or lignite) and light oil, no corrosion problems are anticipated. However, since coal/oil slurry is being handled in this section, erosion of the slurry piping or slurry mix tanks is a possibility. Carbon steel, ASTM A285-C, F, B, Q, is used throughout this section, and an erosion allowance was made in determining the thickness of the vessels and piping. | | | | | |
| 4. Section 3.00-2, Hydrogasification | | | | | |
| a. Stream 306, Steam-Hydrogen Feed to Hydrogasifier | 1200 | 500-1500 | Type-316 Stainless steel | High-temperature steam-hydrogen mixtures | |
| | | | Composition, mole-% | | |
| | | | H ₂ 46.5 | | |
| | | H ₂ O 53.5 | | | |
| | | Total 100.0 | | | |
| b. Stream 309, Gas to Reactor, Second Stage | 1340 | 500-1500 | Refractory | Sulfur components in reducing and carburizing atmosphere at high temperature | |
| | | | Composition, mole-% | | |
| | | | H ₂ 46.5 | | |
| | | H ₂ O 53.5 | | | |
| | | Total 100.0 | | | |
| c. Stream 311, Gas to Reactor First Stage | 1800 | 500-1500 | Refractory | Sulfur components in reducing and carburizing atmosphere at high temperature | |
| | | | Composition, mole-% | | |
| | | | N ₂ 0.27 | | |
| | | | CO 3.21 | | |
| | | | CO ₂ 2.89 | | |
| | | | H ₂ 29.21 | | |
| | | CH ₄ 19.26 | | | |
| | | H ₂ O 44.46 | | | |
| | | H ₂ S 0.70 | | | |
| | | Total 100.00 | | | |

Table 1, Part 3. POTENTIAL CORROSION AREAS IN
HYGAS PILOT PLANT

| Stream/Equipment Description | Temp Range, °F | Conditions Press. Range, psig | Composition, mole % or wt % * | | Material of Construction | Corrosive/Erosive Agents | | | | | |
|--|-------------------|-------------------------------------|--|--------|---|---|----------|-----------------------------|----------------------|-----------------------------------|--|
| | | | | | | | | | | | |
| d. <u>Stream 313</u> , Gas From Reactor Product Separator | 1500 | 500-1500 | N ₂ | 0.45 | Superalloy, Incoloy 800 (Alonized) | Sulfur compounds in reducing and carburizing atmosphere at high temperatures | | | | | |
| | | | CO | 9.84 | | | | | | | |
| | | | CO ₂ | 4.85 | | | | | | | |
| | | | H ₂ | 32.77 | | | | | | | |
| | | | CH ₄ | 17.92 | | | | | | | |
| | | | C ₂ H ₆ | 0.43 | | | | | | | |
| | | | C ₃ H ₈ | 0.16 | | | | | | | |
| | | | H ₂ O | 32.23 | | | | | | | |
| | | | H ₂ S | 1.08 | | | | | | | |
| | | | C ₆ H ₆ | 0.27 | | | | | | | |
| | | | Total | 100.00 | | | | | | | |
| | | | e. <u>Stream 401</u> , Product Gas to Quench Tower | 600 | | | 500-1500 | N ₂ | 0.33 | Low Alloy Steel (ASTM A335-P1) | High-pressure hydrogen and sulfur compounds |
| | | | | | | | | CO | 7.28 | | |
| | | | | | | | | CO ₂ | 3.58 | | |
| H ₂ | 24.22 | | | | | | | | | | |
| CH ₄ | 13.26 | | | | | | | | | | |
| C ₂ H ₆ | 0.32 | | | | | | | | | | |
| C ₃ H ₈ | 0.12 | | | | | | | | | | |
| H ₂ O | 23.83 | | | | | | | | | | |
| H ₂ S | 0.80 | | | | | | | | | | |
| C ₆ H ₆ | 25.70 | | | | | | | | | | |
| NH ₃ | 0.52 | | | | | | | | | | |
| HCN | 0.03 | | | | | | | | | | |
| Total | 100.00 | | | | | | | | | | |
| f. <u>Stream 403</u> , Quench Separator Water to Spent Char Slurry Mix Tank | 200 | 500-1500 | | | Water, saturated with H ₂ S and CO ₂ | | | Carbon steel, (ASTM A53) | Dissolved acid gases | | |
| | | | See Streams 306, 309, 311, 313 and 401, above | | | | | | | | |
| g. <u>Vessel 3.06-01</u> , Hydrogasifier | 600-1800 | 500-1500 | 1. <u>Outer shell</u> : Low- alloy steel (ASTM A515, GR 70 F.B.Q.) except for top 10 feet in char drier section (ASTM A204- GRB, C-1/2 Mo.) | | None, except for char drier section where the atmosphere is high in hydrogen content. | | | | | | |
| | | | 2. <u>Inner shell</u> : Low- alloy steel (ASTM A204- GRB, C-1/2 Mo) lined with 12 in. of light-weight castable, which is, in turn, lined internally with 4 in. of high-alumina castable. | | | | | | | | |
| | | | 3. <u>Internals</u> : The slurry drier cone, product gas separator, first-stage mixing pot ~ Incoloy 800 or RA 330 Stainless Steel. Where possible all internal parts were "Alonized." | | | | | | | | |

**Table 1, Part 4. POTENTIAL CORROSION AREAS IN
HYGAS PILOT PLANT**

| Stream/Equipment Description | Conditions | | Composition mole % or wt % | Material of Construction | Corrosive/Erosive Agents |
|---|-------------------|-----------------------|--|-------------------------------------|--|
| | Temp Range, °F | Press. Range, psig | | | |
| h. Pressure-Equalizing Line Between Hydrogasifier Slurry Drier Section and Steam Separator Drum | 600 | 500-1500 | High-pressure steam and reactor effluent (see stream 401, above) | Low alloy steel (ASTM A-335 Pi) | High-pressure hydrogen and sulfur compounds |
| i. Transfer Line Between Reactor and Spent Char Slurry Mix Drum Conveying Spent Char | 1340 | 500-1500 | High-pressure steam and spent char | Superalloy Incoloy 800 | High-temperature steam and spent char |
| j. Vent From Spent Char Slurry Mix Drum Back to Reactor | 100-1340 | 500-1500 | | Type-316 stainless steel | High-temperature hydrogen |
| k. Hydrogen-Steam Feed Line to Reactor (Not Used With Electrothermal Reactor) | 1200 | 500-1500 | | Type-316 stainless steel (ASTM 312) | High-temperature steam/hydrogen mixtures |
| 5. Section 4.00-1, Product Gas Quench and Separation | | | | | |
| a. Stream 402, Quench Tower Bottoms | 200 | 500-1500 | H ₂ O C ₂ H ₆ Oil Total | Carbon steel (ASTM A53) | |
| b. Stream 405, Quench Water Recycle | 100 | 500-1500 | H ₂ O | Carbon steel (ASTM A53) | |
| c. Stream 406, Quench Tower Off-Gas | 110 | 500-1500 | CO CO ₂ H ₂ H ₂ O CH ₄ C ₂ H ₆ C ₃ H ₈ N ₂ H ₂ S NH ₃ HCN C ₂ H ₄ Total | Carbon steel (ASTM A53) | |
| 6. Section 4.00-2, Product Gas Purification | | | | | |
| a. Stream 408, Absorber Bottoms | 140 | 500-1500 | CO ₂ H ₂ O H ₂ S DGA Total | Carbon steel (ASTM A53) | Aqueous diglycolamine and dissolved acid gases |
| b. Stream 411, Recycled Fresh Amine | 100 | 500-1500 | H ₂ O DGA Total | Carbon steel | Aqueous diglycolamine |

Table 1, Part 5. POTENTIAL CORROSION AREAS IN
HYGAS PILOT PLANT

| Stream/Equipment Description | Conditions | | Composition, mole % or wt % | Material of Construction | Corrosive-Erosive Agents |
|--|-------------------|-----------------------|--|---|---|
| | Temp Range, °F | Press. Range, psig | | | |
| c. <u>Stream 422, Spent Caustic to Disposal</u> | 110 | 500-1500 | CO ₂ 3.82 | Carbon steel (ASTM A53) | Dilute caustic |
| | | | H ₂ O 84.70 | | |
| | | | NaOH 9.42 | | |
| | | | H ₂ S 2.06 | | |
| | | | Total 100.00 | | |
| d. <u>Stream 423, Recycled Caustic Solution</u> | 110 | 500-1500 | CO ₂ 3.83 | Carbon steel (ASTM A53) | Dilute caustic |
| | | | H ₂ O 84.70 | | |
| | | | H ₂ S 2.05 | | |
| | | | NaOH 9.42 | | |
| | | | Total 100.00 | | |
| e. <u>Stream 424, Make-up Caustic</u> | 110 | 500-1500 | H ₂ O 90.00 | Carbon steel | Dilute caustic |
| | | | NaOH 10.00 | | |
| | | | Total 100.00 | | |
| f. <u>Vessel 4.06-02, CO₂ and H₂S Absorber</u> | 110-140 | 500-1500 | See streams 406, 408, and 411, above | Carbon steel | Same as streams indicated, above |
| | | | See streams 423, and 424 above | | |
| g. <u>Vessel 4.06-04, Caustic and Water Wash Scrubber</u> | 100-110 | 500-1500 | See streams 423, and 424 above | Carbon steel | Same as streams indicated, above |
| | | | | | |
| 7. <u>Section 4.00-3, Amine Regeneration</u> | | | | | |
| a. <u>Stream 409, Bottoms from Amine Regenerator</u> | 240 | K | H ₂ O 50.0 | Carbon steel (ASTM A53) | Aqueous diglycolamine |
| | | | NGA 50.0 | | |
| | | | Total 100.0 | | |
| b. <u>Stream 413, Sour Gas</u> | 140 | 2 | CO ₂ 77.56 | Carbon steel (ASTM A53) | Acid gases |
| | | | H ₂ O 9.43 | | |
| | | | H ₂ S 12.00 | | |
| | | | Total 100.00 | | |
| c. <u>Stream 416, Amine to Reboiler</u> | 240 | K | H ₂ O 42.11 | Carbon steel | Aqueous diglycolamine |
| | | | NGA 17.89 | | |
| | | | Total 100.00 | | |
| d. <u>Vessel 4.06-03, Amine Regenerator</u> | 240 | K | See streams 409, 413, and 416 | Carbon steel (A285C, F. B. Q.) | Aqueous diglycolamine and acid gases |
| | | | | | |
| e. <u>Item 4.07-56, Regenerator Reflux Condenser</u> | 140 | K | See streams 408 and 413 | Tubes - S. S. 316 Shell - carbon steel (A 285C, F. B. Q.) | Aqueous diglycolamine and acid gases |
| | | | | | |
| 8. <u>Section 5.00-1, Methanation</u> | | | | | |
| a. <u>Stream 433, First-Stage Feed Gas</u> | 550 | 500-1500 | CO 3.9 | Low-alloy steel (ASTM A204 C, C-1/2 Mo) | High-temperature hydrogen |
| | | | H ₂ 19.3 | | |
| | | | CH ₄ 74.6 | | |
| | | | C ₂ H ₆ 0.2 | | |
| | | | C ₃ H ₈ 0.1 | | |
| | | | N ₂ + C ₆ H ₆ 1.9 | | |
| Total 100.0 | | | | | |

Table 1, Part 6. POTENTIAL CORROSION AREAS IN
HYGAS PILOT PLANT

| Stream/Equipment Description | Conditions | | Composition mole % wt % * | Material of Construction | Corrosive-Erosive Agents |
|---|-------------------|-----------------------|--|---|------------------------------|
| | Temp Range, °F | Press. Range, psig | | | |
| b. <u>Stream 435, First-Stage Effluent</u> | 880 | 500-1500 | H ₂ 7.8 H ₂ O 4.3 CH ₄ 85.8 N ₂ + C ₆ H ₆ 2.1 Total 100.0 | Low-alloy steel (ASTM A204C, C-1/2 Mo) | High-temperature hydrogen |
| c. <u>Stream 437, Second-Stage Feed Gas</u> | 550 | 500-1500 | CO 4.0 H ₂ 19.3 H ₂ O 2.2 CH ₄ 72.3 C ₂ H ₄ 0.2 C ₃ H ₈ 0.1 C ₆ H ₆ + N ₂ 1.9 Total 100.0 | Low-alloy steel (ASTM A204C, C-1/2 Mo) | High-temperature hydrogen |
| d. <u>Stream 434, Effluent From Feed - Effluent Exchanger</u> | 630 | 500-1500 | H ₂ 7.7 H ₂ O 6.7 CH ₄ 83.6 N ₂ + C ₆ H ₆ 2.0 Total 100.0 | Low-alloy steel (ASTM A204C, C-1/2 Mo) | High-temperature hydrogen |
| e. <u>Stream 438, Effluent From Second Stage</u> | 880 | 500-1500 | H ₂ 7.7 H ₂ O 6.7 CH ₄ 83.6 N ₂ + C ₆ H ₆ 2.0 Total 100.0 | Low-alloy steel (ASTM A204C, C-1/2 Mo) | High-temperature hydrogen |
| f. <u>Vessel 5.06-01, First-Stage Methanator and 5.06-02, Second-Stage Methanator</u> | 550-880 | 500-1500 | See streams 433, 435, 437 and 438, above | Low-alloy steel (ASTM A204C, C-1/2 Mo) | High-temperature hydrogen |
| g. <u>Vessel 5.06-41, Methanator Knockout Drum</u> | 100 | 500-1500 | See stream 438, above | Low-alloy steel (ASTM A515, GP-70) | Wet product gas |
| 9. <u>Section 6.00-1, Gas and Liquid Waste Disposal</u> | 100-300 | Ambient to 1500 | See waste streams given in previous sections | Carbon steel | See above |
| 10. <u>Section 6.00-2, Solid Waste</u> | 100-460 | Ambient to 1500 | Char/water slurry | Carbon steel (Tungsten carbide & ceramic inserts used in throttling slurry to minimize wear problems) | Slurry - (abrasive) |
| 11. <u>Section 7.00-1, Hydrogen and Natural Gas Compressor and Superheaters</u> | | | | | |
| a. <u>Stream 304, Hydrogen From Superheater</u> | 1200 | 500-1500 | Process hydrogen | S. S. 316 | High-temperature hydrogen |
| b. <u>Stream 317, Steam From Superheater</u> | 1200 | 500-1500 | Steam | S. S. 316 | High-temperature steam |
| c. <u>Item 7.05-01, Steam Superheater</u> | 1350 | 500-1500 | Steam | Incoloy 800 | High-temperature steam |
| d. <u>Item 7.05-02, Hydrogen Superheater</u> | 1350 | 500-1500 | Process hydrogen | Incoloy 800 | High-temperature hydrogen |

**Table 1, Part 7. POTENTIAL CORROSION AREAS IN
HYGAS PILOT PLANT**

| Stream/Equipment Description | Conditions | | Material of Construction | Corrosive-Erosive Agents |
|---|----------------|-------------------|--|--|
| | Temp Range, °F | Press Range, psig | | |
| 12. Section 7.00-2, Boilers and Air Compressors | | | | |
| No special problems are anticipated in this section of the plant | | | | |
| 13. Section 7.00-3, Water Treatment Facilities | | | | |
| No special problems are anticipated in this section of the plant. The plant cooling water is obtained from the Chicago sanitary drainage canal, however, and must be chlorinated to prevent algae formation and fouling of water-cooled heat exchangers in the plant. | | | | |
| 14. Section 7.22-01, Hydrogen Plant and 7.22-02, Sulfur Plant | | | | |
| These are conventional packaged process plants and materials of construction follow common industry practice. | | | | |
| 15. Section 8.00-1, Electro-thermal Gasification | | | | |
| a. Stream 801, Spent Char From Hydrogasifier | 1400 | 500-1500 | Incoloy 800 (B 407) | Hot char/steam |
| b. Stream 803, Superheated Steam to Electrothermal Gasifier | 1200 | 500-1500 | Type-316 stainless steel (A312) | Superheated steam |
| c. Stream 804, Residue Char to Slurry | 1900 | 500-1500 | Incoloy 800 (B 407) | Hot char |
| d. Stream 806, Char/Steam Mixture to Slurry | 1200 | 500-1500 | Incoloy 800 (B407) | Hot char/steam (corrosive/erosive) |
| e. Stream 807, Electrothermal Gasifier Gas to Hydrogasifier | 1900 | 500-1500 | Incoloy 800 (B407) | High-temperature synthesis gas |
| f. Vessel 8.06-01, Electrothermal Gasifier | 1900 | 500-1500 | Refractory-lined carbon steel shell (electrode S.S. 316) | High-temperature gases and char (corrosive/erosive) |
| g. Item 8.06-44, Char Lift Cyclone | 600 | 500-1500 | Type-316 Stainless Steel | Char/steam mixture (erosive) |
| h. Item 8.06-43, Vent Gas Cyclone | 1900 | 500-1500 | Incoloy 800 | High-temperature synthesis gas and char fine (corrosive/erosive) |

* Mole % for gases and wt % for liquids and solids and for 50:50 design case.
 * Problems have been brought under control by neutralizing acidic liquids.

Table 2. POTENTIAL CORROSION AREAS IN HYGAS PROCESS WITH SYNTHESIS GAS GENERATED BY FLUIDIZED-BED STEAM-OXYGEN GASIFICATION OF RESIDUAL CHAR

| Stream/Equipment Description | Conditions | | Material of Construction | Corrosive/Erosive Agents |
|--|----------------|--------------------|---|--|
| | Temp Range, °F | Press. Range, psig | | |
| The corrosion/erosion problems in most sections of this plant are similar to those listed in Table 1, except for the following additional areas: | | | | |
| 1. Hot Carbonate Scrubbing and Regeneration Sections | 85-140 | 8-1060 | Piping and Vessels: Stress-relieved carbon steel Solutions Pumps, Control Valves, and Reboiler Tubes: 300-Series S. S. | Aqueous potassium carbonate (where H ₂ S is in low concentration film-forming fatty amines should be added as inhibitors. Where H ₂ S is high, corrosion is minimized.) |
| 2. Oxygen-Steam Gasification | | | | |
| a. Oxygen-Steam Gasifier Effluent | 1853 | 1120 | | High-temperature reducing gases containing H ₂ S |
| | | | CO | 15.02 |
| | | | CO ₂ | 15.02 |
| | | | H ₂ | 23.27 |
| | | | H ₂ O | 43.21 |
| | | | CH ₄ | 3.36 |
| | | | H ₂ S | 0.12 |
| | | | Total | 100.00 |
| b. Low-temperature Zone Gasifier Effluent | 1273 | 1100 | | High-temperature reducing gases containing H ₂ S |
| | | | CO | 18.00 |
| | | | CO ₂ | 18.53 |
| | | | H ₂ | 22.84 |
| | | | H ₂ O | 24.35 |
| | | | CH ₄ | 14.07 |
| | | | C ₂ H ₆ | 0.53 |
| | | | N ₂ | 0.04 |
| | | | NH ₃ | 0.40 |
| | | | H ₂ S | 0.90 |
| | | | C ₆ H ₆ | 0.17 |
| | | | Oil | 0.17 |
| | | | Total | 100.00 |

Because of differences in the process flowsheet, the following sections are peculiar to the HYGAS pilot plant and are either different or do not appear in this variation of the HYGAS process:

1.) Section 3.00-1, Reactor Feed Slurry Preparation; 2.) Section 4.00-2, Product Gas Purification; 3.) Section 4.00-3, Amine Regeneration. In this process variation coal feed is introduced via lock hoppers and gas purification is by hot carbonate scrubbing rather than amine scrubbing.

* Mole % for gases, wt % for liquids and solids

Table 3, Part 1. POTENTIAL CORROSION AREAS IN HYGAS DEMONSTRATION PLANT

| Steam/Equipment Description | Conditions | | Material of Construction | Corrosive/Erosive Agents |
|---|----------------|--------------------|--------------------------|--|
| | Temp Range, °F | Press. Range, psig | | |
| 1. Unit 2300, Selective Potash Scrub | | | | |
| a. Stream 2311, Lean Solution | 105 | 1150 | -- | Aqueous KOH containing dissolved acidic and basic gases. |
| | | | | |
| b. Stream 2312, Rich Solution | 145 | 1150 | -- | |
| c. Stream 2313, Sour Condensate | 100 | 1150 | -- | |
| 2. Unit 2600, Hot Potash Scrub | | | | |
| a. Stream 2605, Lean Solution | 190 | 5 | -- | Aqueous KOH containing dissolved acidic gases |
| | | | | |
| b. Stream 2606, Semi-Lean Solution | 200 | 5 | -- | Aqueous KOH containing dissolved acidic gases |
| | | | | |
| c. Stream 2607, Rich Solution | 219 | 5 | -- | Aqueous KOH containing dissolved acidic gases |
| 3. Unit 3200, Sulfur Recovery | | | | |
| a. Stream 2303, Acid Gas From Unit 2300 | 125 | 6.3 | -- | Moist acidic and basic gases |

The corrosion-erosion problems in most sections of this plant are similar to those listed in Table 1, except for the following additional areas:

| Stream/Equipment Description | Temp Range, °F | Press. Range, psig | Composition, mole % or wt % * | Material of Construction | Corrosive/Erosive Agents |
|---|----------------|--------------------|--|--------------------------|--|
| 1. Unit 2300, Selective Potash Scrub | | | | | |
| a. Stream 2311, Lean Solution | 105 | 1150 | CO ₂ ** H ₂ S HCN COS + CS ₂ NH ₃ KOH H ₂ O | -- | Aqueous KOH containing dissolved acidic and basic gases. |
| b. Stream 2312, Rich Solution | 145 | 1150 | ** | -- | |
| c. Stream 2313, Sour Condensate | 100 | 1150 | ** | -- | |
| 2. Unit 2600, Hot Potash Scrub | | | | | |
| a. Stream 2605, Lean Solution | 190 | 5 | CO ₂ 9.11 H ₂ O 65.49 KOH 22.58 Formate 2.82 Total 100.00 | -- | Aqueous KOH containing dissolved acidic gases |
| b. Stream 2606, Semi-Lean Solution | 200 | 5 | CO ₂ 9.45 H ₂ O 65.25 KOH 22.49 Formate 2.81 Total 100.00 | -- | Aqueous KOH containing dissolved acidic gases |
| c. Stream 2607, Rich Solution | 219 | 5 | CO ₂ 12.89 H ₂ S 0.01 H ₂ O 62.55 KOH 21.82 Formate 2.73 Total 100.00 | -- | Aqueous KOH containing dissolved acidic gases |
| 3. Unit 3200, Sulfur Recovery | | | | | |
| a. Stream 2303, Acid Gas From Unit 2300 | 125 | 6.3 | H ₂ S 16.23 CO ₂ 73.09 NH ₃ 0.01 HCN 0.39 CO 0.44 H ₂ 0.44 CH ₄ 1.10 C ₂ H ₆ 0.04 H ₂ O 8.26 Total 100.00 | -- | Moist acidic and basic gases |

Table 3, Part 2. POTENTIAL CORROSION AREAS IN
HYGAS DEMONSTRATION PLANT

| Steam/Equipment Description | Conditions | | | Material of Construction | Corrosive/Erosive Agents |
|---|----------------|--------------------|---|--------------------------|------------------------------|
| | Temp Range, °F | Press. Range, psig | Composition, mole for wt % | | |
| b. <u>Stream 3202</u> , Acid Gas | 125 | 6.3 | H ₂ S 16.23 CO ₂ 73.09 NH ₃ 0.01 HCN 0.39 CO 0.44 H ₂ 0.44 CH ₄ 1.10 C ₂ H ₆ 0.04 H ₂ O 8.26 Total 100.00 | -- | Moist acidic and basic gases |
| c. <u>Stream 3203</u> , Bypass Gas | 125 | 4.5 | H ₂ S 16.23 CO ₂ 73.09 NH ₃ 0.01 HCN 0.39 CO 0.44 H ₂ 0.44 CH ₄ 1.10 C ₂ H ₆ 0.04 H ₂ O 8.26 Total 100.00 | -- | Moist acidic and basic gases |
| d. <u>Stream 3102</u> , Acid Gas From Unit 3100 | 150 | 6.3 | H ₂ S 16.28 CO ₂ 58.42 NH ₃ 3.34 HCN 1.09 H ₂ O 20.87 Total 100.00 | -- | Moist acidic and basic gases |
| e. <u>Stream 3206</u> , Acid Gas to Furnace | 350 | 6.0 | H ₂ S 16.25 CO ₂ 67.38 NH ₃ 1.31 HCN 0.66 CO 0.27 H ₂ 0.27 CH ₄ 0.67 C ₂ H ₆ 0.02 H ₂ O 13.17 Total 100.00 | -- | Moist acidic and basic gases |

Table 3, Part 3. POTENTIAL CORROSION AREAS IN
HYGAS DEMONSTRATION PLANT

| Stream/Equipment Description | Temp Range, °F | Press. Range, psig | Composition, mole for wt % * | Material of Construction | Corrosive/Erosive Agents |
|--------------------------------|-------------------|-----------------------|--|-----------------------------|---------------------------------|
| f. Stream 3215, Claus Tail Gas | 305 | 0.7 | H ₂ S 0.54 SO ₂ 0.27 S ₈ 0.01 S ₂ 0.02 Liquid S 0.03 COS 0.01 CS ₂ 0.01 CO ₂ 49.64 NH ₃ 0.004 HCN 0.17 CO 0.19 H ₂ 0.19 CH ₄ 0.47 C ₂ H ₆ 0.02 H ₂ O 20.35 N ₂ 28.08 Total 100.00 | -- | Moist acidic and basic gases |

* Mole % for gases, wt % for liquids and solids

** Unavailable, proprietary information

PART IV

HYDROGEN GENERATION

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- 4a.2 Electrothermal/2.25 MW (30-Inch) Unit
- 4b Steam-Oxygen Gasification
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4.0 Summary of Hydrogen Generation

The HYGAS process for the hydrogasification of coal is tailored to maximize the direct production of methane in the hydrogasification reactor by the use of high temperature (1200°-1700°F) and high pressure (1000 psig). The process can use as feed caking bituminous coals as well as noncaking lignite and subbituminous coals.

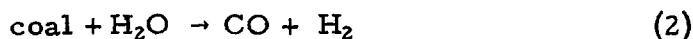
The HYGAS process is based on gasification studies that started at the Institute of Gas Technology (IGT) in 1944. By the mid-1950's, two processes to convert coal to synthetic pipeline gas were being developed: 1) gasification of powdered coal in suspension with oxygen and steam to produce a mixture of carbon monoxide and hydrogen (synthesis gas), which was then methanated, and 2) direct hydrogenation of the coal at elevated temperature and pressure in a fluidized bed. The present HYGAS process incorporates the principles developed in both of these concepts.

Of the total methane formed in the HYGAS process, 58 to 78% is directly formed in the hydrogasifier. This is a key feature of the process, and contributes significantly to the high overall thermal efficiency of more than 70% (when steam-iron is used). If the process were operated at a lower pressure, more methane would be made indirectly, and losses in efficiency would result.

The second key feature of the process is the use of hydrogen and steam in hydrogasification. The coal-hydrogen reaction,



is strongly exothermic, while the coal-steam reaction,



is strongly endothermic. By using a mixture of hydrogen and steam instead of hydrogen alone, the heat released by Reaction 1 is absorbed in situ by Reaction 2, resulting in

- Built-in temperature control, and
- Internal hydrogen generation.

The raw gas from the hydrogasifier contains carbon monoxide and hydrogen. These are converted to methane through Reaction 3,



in the catalytic clean-up methanation unit, where the carbon monoxide content is reduced to the pipeline standard of less than 0.1%, and the heating value of the product gas is raised to satisfactory SNG levels.

The HYGAS pilot plant, completed in the spring of 1971, is designed to deliver 1.5 million SCF of SNG daily from 75 tons of coal feed. In the early spring of 1973,** large-scale conversion of coal to pipeline-quality gas was demonstrated for the first time. In the spring of 1974,* IGT completed a run during which the plant was in operation for over 27 days. During this run, about two-thirds of all the methane created was produced directly in the hydrogasifier, confirming predictions; reaction rates and thermal effects were as expected.

Plant modifications were made during 1974 to study the integration into the base of the pilot plant hydrogasifier reactor of a steam-oxygen gasifier for the production of hydrogen-rich gas from hydrogasified coal char. Design and construction of a steam-iron system was begun by IGT under a separate OCR contract* during 1973 at the IGT pilot plant site. The electro-thermal gasification of char was successfully demonstrated in batch tests; that part of the pilot plant was then put on standby. Utilizing a single SNG plant, the three hydrogen-rich gas-producing facilities could provide a means for close comparative study of hydrogen supply techniques.

4.0.1 Hydrogasifier Reactor

In the pilot plant, the hydrogasifier reactor vessel, with four internally connected contact stages, is 135 feet high and has a 5.5-foot inside diameter. The slurry is sprayed into the top stage, a 2.5-foot-diameter, 10-foot-high fluidized drying bed. The sensible heat in the gaseous reaction products from the later stages vaporized the oil. At this point, the dry coal is heated to about 600°F.

The coal flows by gravity from the drying bed into a 3-inch-diameter, vertical-lift-line reactor in which the hot gases (1700°F) from the reaction section below provide the lifting force, the heat to raise the solids temperature to 1200°F, and hydrogen that reacts with about 20% of the coal to produce methane. This is the first stage of hydrogasification. At the top of the lift line, the gas and coal disengage. The gas moves up to vaporize the oil in the slurry-drying stage. The partially reacted coal can be split into two streams. Part of it can be transferred to the base of the lift line to be mixed with the incoming fresh coal. By this means, it is expected to feed raw caking coal directly to the reactor without pretreatment. Eliminating pretreatment can reduce gas cost significantly.

The remainder of the partially reacted coal flows by gravity to the second-stage hydrogasifier. Here the solids are heated in a fluidized bed to about 1700°F and further gasified by the steam and hydrogen-rich gas rising from the steam-oxygen gasification stage below. (Or, alternatively, the hydrogen-rich gas may flow from the steam-iron reactor or the electro-thermal generator.) The second-stage bed is 2.5 feet in diameter, is lined with refractory, and is 15 feet deep.

In this second stage of the reactor, the exothermic hydrogen reaction produces methane, and the endothermic steam reaction produces carbon monoxide and hydrogen. If the temperature rises, the steam-char reaction

* OCR Contract No. 14-32-0001-1518; now ERDA Contract No. E(49-18)1518.

** Following the contract period reported here.

speeds up and prevents the temperature from rising any further. If the temperature drops, the steam-char reaction slows down and thus provides an automatic temperature control. About 25% more of the coal is converted in this reaction stage, making the total about 45% in the two stages.

From this reaction stage, the char goes to the hydrogen-producer stage, where, depending on the process being used, the char undergoes different degrees of additional gasification. The steam-oxygen gasifier being used as the hydrogen producer in the pilot plant is directly below the second-stage hydrogasifier. The steam and high-purity oxygen introduced into the gasifier convert char into hydrogen and carbon oxides at 1850°F in a fluidized bed 2 feet in diameter and 12 feet deep. Ash is discharged from this stage without being slagged. The ash is discharged into a tank where water is added to make a slurry, which is then depressurized. The ash is recovered by filtering and the water used again.

4.0.2 Gas Purification

The composition of the gas leaving the slurry dryer at the top of the hydrogasification reactor depends on the type of hydrogen producer, as shown in Table 4-1. In addition to these major components, the gas contains the slurry oil, coal dust, and trace constituents such as ammonia and hydrogen cyanide.

Table 4-1. GAS COMPOSITION LEAVING THE HYDRO-GASIFICATION REACTOR (Oil-Free) FOR THREE HYDROGEN SUPPLY SYSTEMS

| <u>Component</u> | <u>Electrothermal</u> | <u>Steam-Oxygen</u> | <u>Steam-Iron</u> |
|-------------------------------|-----------------------|---------------------|-------------------|
| CO | 21.3 | 18.0 | 7.4 |
| CO ₂ | 14.4 | 18.5 | 7.1 |
| H ₂ | 24.2 | 22.8 | 22.5 |
| H ₂ O | 17.1 | 24.4 | 32.9 |
| CH ₄ | 19.9 | 14.1 | 26.2 |
| C ₂ H ₆ | 0.8 | 0.5 | 1.0 |
| H ₂ S | 1.3 | 0.9 | 1.5 |
| Other | 1.0 | 0.8 | 1.4 |
| Total | 100.0 | 100.0 | 100.0 |

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In the pilot plant, this mixture at 600°F passes to a baffle tower in which it is quenched and washed with water. This removes the dust and water-soluble trace components and condenses the water and light-oil vapors. The gas then flows to a conventional, packed-tower acid-gas removal system in which the carbon dioxide and the hydrogen sulfide are absorbed in a diglycolamine-water solution. Upon regenerating this solution, the carbon dioxide

and hydrogen sulfide are released and flow to a Claus plant for sulfur recovery. The amine purification system used in the pilot plant is not intended as a commercial design because it does not provide for separate collection of the various constituents. It was selected because it can handle the wide range of acid-gas concentrations arising from the various coals to be tested.

4.0.3 Methanation

As it enters the methanation section, the purified gas would typically have the composition shown in Table 4-2. The methanation step has two purposes: One is to raise the heating value of the gas to near that of methane; the other is to reduce the carbon monoxide concentration to the requisite 0.1% or less. This is accomplished by carrying out Reaction 3 given earlier.

Table 4-2. GAS COMPOSITION ENTERING THE METHANATOR, AS CALCULATED FOR THREE HYDROGEN SUPPLY SYSTEMS

| <u>Component</u> | <u>Electrothermal</u> | <u>Steam-Oxygen</u> | <u>Steam-Iron</u> |
|-------------------------------|-----------------------|---------------------|-------------------|
| CO | 16.8 | 18.0 | 12.8 |
| CO ₂ | 2.0 | 2.0 | 2.0 |
| H ₂ | 50.5 | 54.0 | 38.5 |
| CH ₄ | 29.5 | 25.0 | 45.0 |
| C ₂ H ₆ | 1.2 | 1.0 | 1.7 |
| Total | 100.0 | 100.0 | 100.0 |

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To obtain nearly complete elimination of carbon monoxide and low residual hydrogen (pipeline-quality gas) in the methanation section, the ratio of hydrogen to carbon monoxide is adjusted, usually by a water-gas shift reaction, to slightly above 3. High pressure and low temperature favor completion of the methanation reaction. Very reactive, high-nickel-content catalysts are generally preferred to make the reaction proceed rapidly at the low temperatures employed. The temperature of the catalyst must be above 450 °F to avoid formation of nickel carbonyl, which causes depletion of the nickel content of the catalyst, and below about 950 °F to avoid carbon deposition and catalyst sintering. The reaction is very exothermic; therefore, to avoid excessive temperature rises, the HYGAS Process uses a cold-gas recycle system.

4.0.4 Hydrogen for the HYGAS Process

The HYGAS process is very flexible in that it can successfully use any source of hydrogen. The steam-oxygen version employs an oxygen system, whereas the electrothermal and steam-iron concepts are air-based. The manner in which each hydrogen system affects overall process material balances is indicated in Figure 4-1.

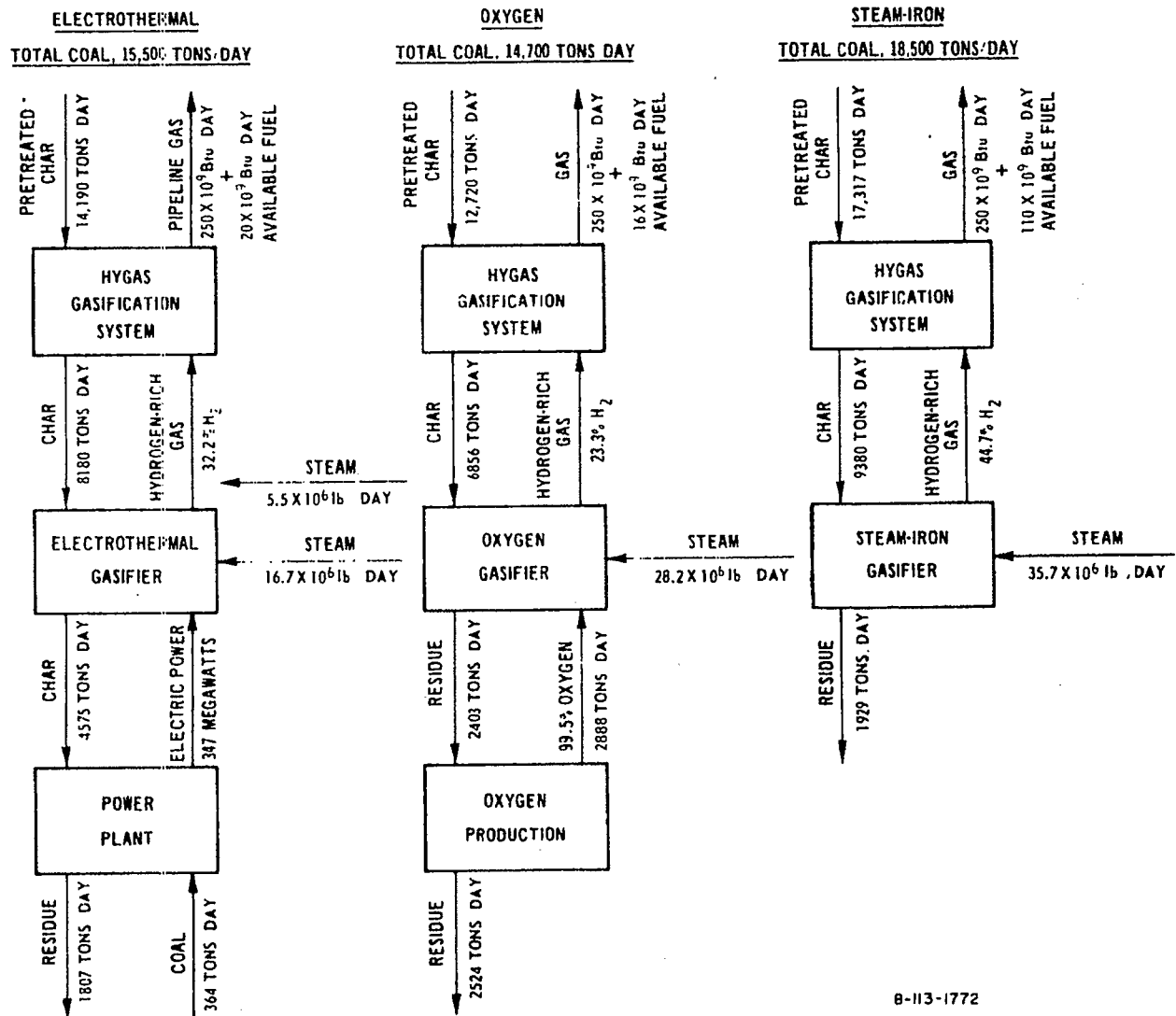


Figure 4-1. EFFECT ON THE IGT HYGAS PROCESS FROM USING THREE DIFFERENT SYSTEMS FOR PRODUCING HYDROGEN-RICH GAS: PLANT CAPACITY IS 250 X 10⁹ Btu/DAY OF PIPELINE-QUALITY GAS

4.0.4.1 Electrothermal/300 kW (6-Inch) Unit

The HYGAS program concept of a fully integrated hydrogasification plant for the conversion of coal to pipeline gas led to a search for a technically feasible and economically attractive source of hydrogen. After reviewing several processes in the advanced stages of development, HYGAS personnel, with the concurrence of OCR, decided to initially investigate the electrothermal gasification approach. A fluidized bed of spent hydrogasified char, internally heated by an immersed electrode or electrodes, would be made to react with steam to produce a hydrogen-carbon monoxide rich synthesis gas. This gas could be directly fed to the hydrogasifier, thus utilizing the sensible heat of both the product gas and hydrogasified char while also eliminating costly clean-up and compression stages (see Figure 4-2).

Technological advances made in recent years by investigators of electrothermal fluidized-bed systems had given promise that a pilot unit could be designed, built, and operated successfully. Since the first commercial use of the technique for the production of hydrocyanic acid¹, processes for the calcination of petroleum coke² and various metallurgical applications³ have emerged and prospects for future developments are numerous.^{3, 4}

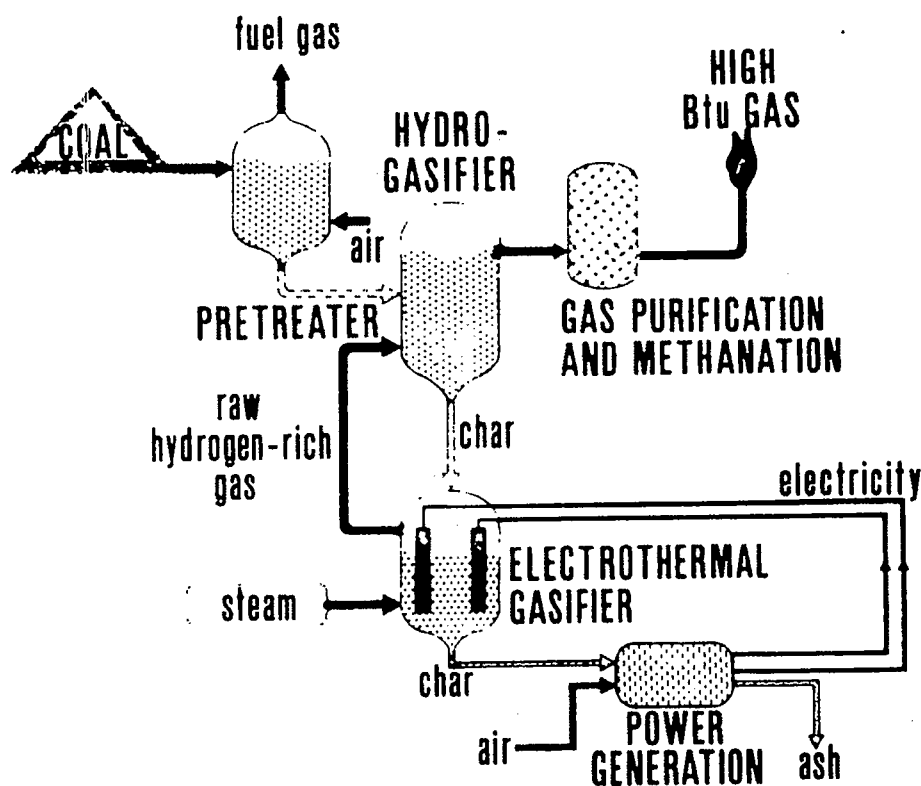
Preliminary studies^{5, 6} were performed for design purposes using spent hydrogasified char in an electrothermal fluidized bed to compare data with the published results for the various carbonaceous materials (graphite, petroleum coke, etc.) generally investigated. Results of these studies showed that the electrical characteristics of a spent char system are similar to those of the other materials^{7, 8, 9} and that a low-voltage and high-amperage operation could be expected. With the information available, a 300-kilowatt electrothermal pilot unit was designed and constructed. It was a continuous system capable of operating at pressures of 1250 pounds per square inch (gage), reactor temperatures of 2100°F, char feed rates to 350 pounds per hour, and steam feed rates of 300 pounds per hour. Major areas investigated were reaction rates, voltage-current relationships at various power input levels, electrode configuration, development of nonconsumable electrodes, and general system operability. The establishing of these criteria led to the design of a 2.25 megawatt system for integration into the HYGAS pilot plant.

4.0.4.2 Electrothermal/2.25 MW (30-Inch) Unit

The 2.25-MW electrothermal gasifier was built and operated successfully. Its diameter inside the refractory lining is 30 inches, and the reactor height is 50 feet. The electrode configuration is concentric. The gasifier has automatic and manual modes for controlling power input.

The system piping has been successfully leak-tested to design pressures, instrumentation has been loop-checked, and shakedown of the electrical control system was undergoing shakedown at termination of the contract period reported here. Additional information concerning this work may be found in Interim Report No. 1 on the HYGAS Process, OCR Contract No. 14-31-0001-1221 (now E(49-18)-1221, under ERDA), OCR R&D Report No. 110.

In the years since the development of this method was begun, the cost of electric power has risen sharply; therefore, this method is now the least economical of the three.



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Figure 4-2. THE HYGAS PROCESS SHOWN WITH AN ELECTROTHERMAL GASIFIER TO PRODUCE HYDROGEN-RICH GAS

4. 0. 4. 3 Steam-Oxygen Gasification

The objective of the steam-oxygen gasification program was to investigate the feasibility of, and to develop an oxygen-based synthesis-gas generator that would provide a hydrogen-bearing gas for use in the HYGAS hydrogasification process for conversion of coal to pipeline quality SNG.

Since 1964, the Institute of Gas Technology has been engaged in the development of an integrated process for the conversion of coal to pipeline-quality gas. In developing an integrated process concept, investigators have been constantly aware of the major effect the cost of hydrogen generation has on the overall cost of producing SNG. In this work, a number of approaches have been explored at IGT for the generation of hydrogen using spent hydrogasifier char as the raw material.

Production of hydrogen from coal or partially spent char is essential in coal-to-gas conversion. More than 1 cubic foot of hydrogen or hydrogen equivalent is required for each cubic foot of methane produced.

Success in using a synthesis gas-steam mixture for hydrogasification in place of a prepurified hydrogen-steam mixture encouraged re-investigation of the potential use of oxygen as a means to support the steam-carbon reaction. In this study, char was partially oxidized with oxygen in the presence of steam at temperatures of 1850° and 1650°F. The latter is as low a temperature as investigators believe might ever prove practical; work at 1650°F was included to indicate the minimum oxygen requirement.

A 4-inch-diameter fluidized-bed balanced-pressure reactor was chosen for the work. In the reactor, an oxygen-steam mixture is used to fluidize spent char from the hydrogasifier. Char combustion to CO and CO₂ and methane formation supply the heat for the steam-char reaction that produces H₂ and CO. Methane formation was assumed to occur in an amount that would be produced at equilibrium. Heat generated by this methane formation is extremely important in minimizing oxygen requirements. Experiments had indicated that amounts in excess of gas-phase equilibrium can occur under some circumstances.

Economics and the conservation of resources by high utilization are the principal factors in ultimately deciding whether an electrothermal, oxygen or steam-iron system will be used to generate the hydrogen-rich gas required for the HYGAS or other hydrogasification processes. Because oxygen addition results in higher carbon oxide levels, the hydrogasifier reactor volume will be somewhat larger than needed with the use of an electrothermal gasifier at an equivalent operating pressure, for equal-volume production of SNG. A larger scrubbing system — again, on an equivalent-production basis — would also be required with an oxygen system to remove the additional carbon oxides formed. The incremental increases in vessel sizes and system duties required for oxygen and electrothermal systems, however, may be more than offset by the savings gained by using oxygen. Although a process may be developed for complete consumption of the carbon in the gasifier, it may be desirable to undertake only partial conversion of the char. The residual char would then be used as a fuel for steam generation.

A major task was the investigation of a specific temperature range that must be used in the gasifier to enable the reactions to proceed at reasonable rates yet allow for maximum methane formation. IGT investigators believe that 1650°F may be too low a temperature to allow practical-sized equipment when using bituminous coal, although 1650°F may be a realistic temperature with lignite or subbituminous coal. An 1850°F gasifier temperature should be quite practical from a kinetic standpoint, although — as the gasifier temperature is increased — the potential increases for problems associated with ash softening. However, in a properly designed system, a temperature of 1850°F should not present problems with most coals.

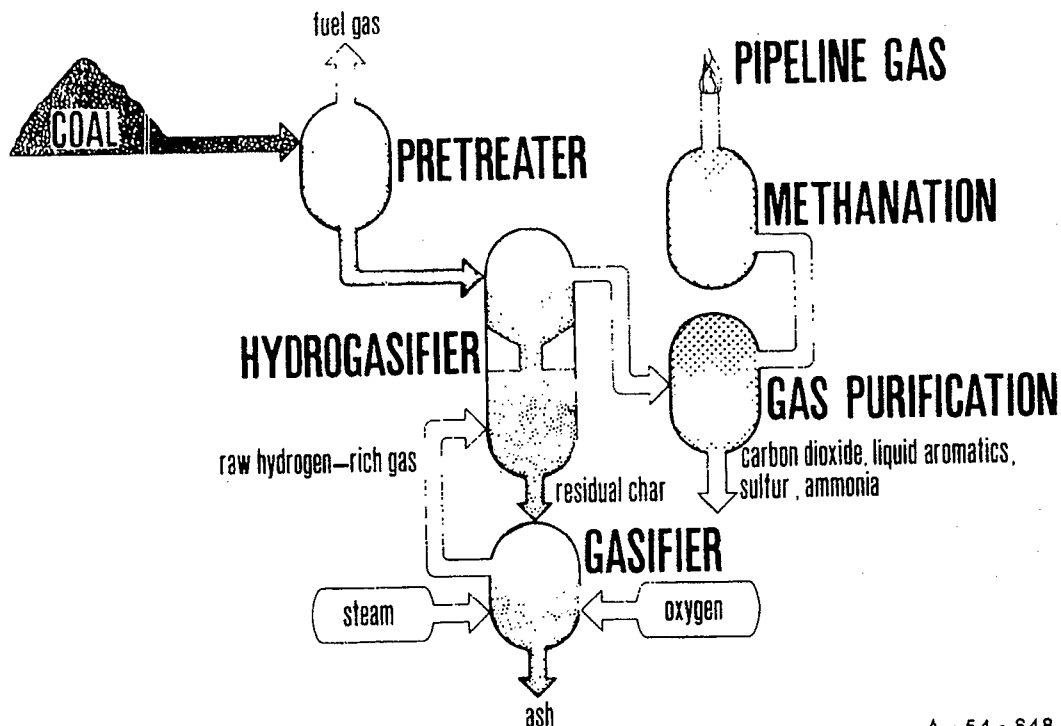
The basic questions to be resolved in the steam-oxygen program are —

- What is the necessary size of the oxygen-steam gasification reactor?
- What is the necessary size of the hydrogasifier when using synthesis gas produced in the oxygen-based gasifier?

Investigators determined that char space velocity and char residence time data obtained in the fluidized-bed gasifier development tests performed in the 6-inch-diameter reactor may be applied toward estimating the size of the steam-oxygen gasifier reactor. Kinetic equations to predict rates of carbon gasification in synthesis gas mixtures were developed from data obtained in the thermobalance studies. These equations and rates are a basis for determining hydrogasifier size when using synthesis gas produced in the steam-oxygen gasifier. The final resolution of the basic questions stated above, however, will be dependent upon data obtained from the HYGAS pilot plant reactor operating with the steam-oxygen gasifier on an integrated basis.

The steam-oxygen gasifier (Figure 4-3) produces a hydrogen-rich gas by reacting the hot char from the hydrogasifier with steam and oxygen in a high-pressure fluidized bed. An operating temperature of 1800°-1900° F is maintained by controlling the quantities of steam and oxygen.

Multiple-feed gas cones inject premixed steam and oxygen at the bottom of the bed. Nonslagging conditions are maintained by a high fluidizing velocity at the reactor bottom and by the gas inlet design, which avoids stagnant areas and promotes rapid initial contact of oxygen, steam, and char. Ash is discharged through the space between the cones.



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Figure 4-3. THE HYGAS PROCESS SHOWN WITH A STEAM-OXYGEN GASIFIER TO PRODUCE HYDROGEN-RICH GAS

The hydrogen content of the gas from this gasifier is the lowest of the three systems. The mechanical simplicity of this system is partially offset by the need for an oxygen plant, the necessity of removing additional carbon dioxide, and the requirement for a longer solids residence time in the hydrogasifier.

Encouraged by the results of this work, pilot-scale verification has been planned at the HYGAS pilot plant. During the spring of 1974, a steam-oxygen char gasifier was installed in the base of the HYGAS pilot plant reactor, replacing the original heat-exchange bed that occupied that space. Details of this development are contained in Interim Report No. 1 on the HYGAS process, OCR Contract No. 14-32-0001-1221 (now E(49-18)-1221, under ERDA), OCR R&D Report No. 110.

4.0.4.4 Steam-Iron Gasification

The steam-iron process was considered as a means to provide hydrogen-rich gas for the initial operations of the HYGAS pilot plant. At that time (1969), IGT had been engaged in privately-funded proprietary research and development concerning steam-iron processes for the past eight years. Protracted negotiation of licensing arrangements with assignees of patents derived from this work prohibited an earlier start on the design and construction of process development hardware for making hydrogen-rich gas to be used in pilot-synthesis of SNG.

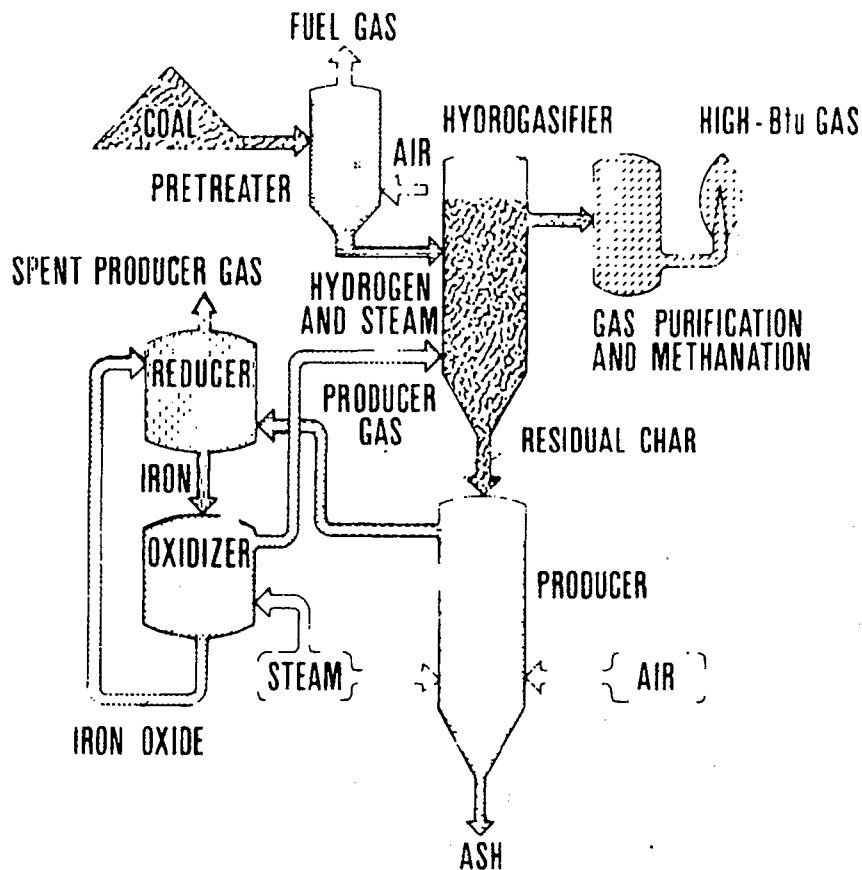
The steam-iron process for making hydrogen is very old. Historically, the early process was operated at atmospheric pressure using two beds of iron solids. One bed was reduced from iron oxide to iron by a suitable gas while the second bed was being oxidized by steam from iron to iron oxide.

Upon oxidizing the iron, the steam was converted to hydrogen. When the beds were fully reacted, valves switched the gases from one bed to the other and the reverse operation was carried out. This process has been largely abandoned in favor of other hydrogen processes, especially steam-reforming of natural gas.

The new IGT process replaces the cyclic operation with a continuously flowing system that utilized fluidized-bed reactors. The process operates at HYGAS pressure so that the hydrogen-steam mixture produced can be passed directly into the hydrogasification reactor. The steam-iron version of the HYGAS Process, shown in Figure 4-4 is quite complex compared with the other versions. The process is potentially superior in efficiency, however, and in the cost of the gas produced.

Spent char from the hydrogasifier would be fed directly to the producer vessel in which the char reacts with air and steam to generate a gas capable of reducing iron oxide to iron. Operation of the producer at temperatures near 2000°F should yield good reducing gas having CO/CO₂ and H₂/H₂O ratios exceeding 4:1. The hot reducing gases are fed directly to the reducer where they contact a recirculating stream of iron oxide and reduce it. The reduced oxide is contacted with steam in the oxidizer and reoxidized, producing a mixture of hydrogen and excess steam.

The reduction-oxidation reactor will be designed with four fluidized-bed stages, two each in the reducer and the oxidizer. The double beds ensure the highest steam and reducing-gas conversions for the operating conditions used. Locating the reducer on top of the oxidizer allows the spent reducing gases to convey the iron solids to the top of the reactor. The new steam-iron process is especially suited to the HYGAS Process because the product gas from the oxidizer can be fed directly at temperature and pressure into the second-stage hydrogasification reactor. Because the hydrogasification reactor operates well with a 60:40 steam/hydrogen ratio, it is not necessary to achieve high steam conversion in the steam-iron process. The very low carbon oxide content in the raw gas from the steam-iron reactor results in a very low carbon oxide content in the raw gas from the hydrogasifier. This, combined with the very high hydrogen content, permits a small hydrogasifier reactor size and much reduced scrubbing requirements.



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Figure 4-4. THE HYGAS PROCESS SHOWN WITH A STEAM-IRON SYSTEM TO PRODUCE HYDROGEN-RICH GAS

The steam-iron section provides orientation concerning how the steam-iron process would supplement a hydrogasification process. In 1974, as this report is prepared, the steam-iron process is under active development for application in hydrogasification, under the 1973 OCR Contract No. 14-32-0001-1518 (now ERDA Contract No. E(49-18)-1518). Details of the steam-iron system now under development at IGT will be presented in full at the appropriate time when the development is more advanced, in accordance with ERDA requirements.

Details of all steam-iron contract work to date — including process flow sheets — may be found in the U.S. Department of the Interior, Office of Coal Research, R&D Report No. 95, Interim Report No. 1, Development of the Steam-Iron System for Production of Hydrogen for the HYGAS Process, Washington, D.C. 1974.

IGT has been working since 1961 on various approaches to steam-iron gasification for use in the direct synthesis of SNG. The earlier IGT work was sponsored by Fuel Gas Associates (Consolidated Natural Gas Service Company, Inc., Texas Eastern Transmission Corp., and Consolidation Coal Company). More recently the work has been sponsored by the American Gas Association and OCR.

Various steam-iron process patents included here as Appendix 4c-A illustrate the scope of the early, private steam-iron R&D work at IGT and the platform of expertise upon which the IGT-OCR contract cited above is based. All of the patents of Appendix 4c-A are assigned to Consolidation Coal Co., a member of the sponsoring Fuel Gas Associates* (FGA).

The three hydrogen-rich gas producing processes briefly described in this summary are discussed in detail in the body of Part IV of this report.

* In subsequent OCR negotiations after the period reported here (but prior to the establishment of ERDA), FGA — through the American Gas Association — entered into an agreement with OCR assigning limited rights to certain FGA steam-iron patents for the sole purpose of hydrogen-rich gas production to convert coal to SNG. This limitation remains in effect in mid-1975 as this report goes to press.

4.0.5 References Cited

1. Johnson, H. S. and Anderson, A. H., U.S. Patent 2,958,584 (November 1, 1960).
2. Paquet, J. L. and Foulkes, P. B., Can. J. Chem. Eng. 43, 94 (1965).
3. Goldberger, W. M., Hanway, J. E., Jr. and Langston, B. G., Chem. Eng. Progr. 61, 63 (1965).
4. Johnson, J. S., Can. J. Chem. Eng. 39, 145 (1961).
5. Lee, B. S., Pyrcioch, E. J. and Schora, F. C. Jr., "Electrical Resistivity of a High-Pressure Fluidized Bed," Preprint 29B 61st Annual Meeting of The American Institute of Chemical Engineers, Los Angeles (1968) December 1-5.
6. Iowa State University, 2nd Quarterly Progress Report on Contract No. 14-01-0001-479, Dept. of the Interior, Office of Coal Research, Washington, D. C. (1968).
7. Graham, W. and Harvey, E. A., Can. J. Chem. Eng. 43, 146 (1965).
8. Graham, W. and Harvey, E. A., Can. J. Chem. Eng. 44, 17 (1966).
9. Reed, A. K. and Goldberger, W. M., Chem. Eng. Progr. Symp. Ser. 62, 71 (1966).

PART 4a.1

Electrothermal/300 kW (6-Inch) Unit

4a-1-i

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4a.1-1

CHARACTERISTICS OF VARIOUS FEED CHARS

4a.1-22

4a.1.0 Summary of Electrothermal/300 kW (6-Inch) Unit

A summary of this work appears in section 4.0.

4a.1.1 Introduction

A number of studies have been made on the electrical characteristics of fluidized beds of coke or graphite particles; however, none of the studies were carried out at high pressure. Little is reported on the phenomenon of high-pressure fluidization. The effect of pressure on the electrical behavior of fluidized beds, together with fluidization characteristics at high pressure had been studied in the 4-inch-diameter hydrogasification development unit¹. The results of this study were applied to the design of the 6-inch electrothermal gasifier.

The fluidized bed material was a char produced from gasifying a bituminous coal. Average particle size of the char was 240 microns. The fluidizing medium was nitrogen admixed with steam.

The electrical resistivity in a 4-inch fluidized bed of coal char was found to be essentially constant with current density at ambient pressures, but decreased with increasing current density at high pressures.

The data taken at four different pressures are shown in Figures 4a.1-1, -2, -3, and -4. Current density is calculated on the basis of the immersed surface area of the center electrode. Resistivity, ρ , is calculated according to the basic definition:

$$R = \frac{\rho l}{A} \quad (1)$$

where

R = overall resistance between the electrodes

l = distance between electrodes

A = mean surface area of electrodes

For a cylindrical geometry, between two cylinders at radii r and $r + dr$ with potential difference of dV , based on the above definition of resistance, the current flow, I , for an electrode immersion depth L , is -

$$I = \frac{-dV}{\frac{\rho dr}{2\pi rL}} \quad (2)$$

Integrating this expression between limits of the center electrode radius and reactor inside radius, r_e and r_i , and substituting, by definition, $R = \frac{V}{I}$, we obtain -

$$\rho = \frac{2\pi LR}{\ln \frac{r_i}{r_e}} \quad (3)$$

Bed resistivity in all instances decreased with increasing current density; i. e., a nonlinear voltage-current relationship was demonstrated. Reed and Goldberger² observed this trend at ambient pressure at current densities above 0.3 amperes per square inch, but noted a constant resistivity at current densities below 0.3. These authors noticed the presence of small arcs within the bed above 2 amperes per square inch, indicating to them the onset of gas conduction. By this reasoning, even though the bed could not be observed in early HYGAS studies mentioned, the decreasing resistivity should lead to the conclusion of arcing in the bed. However, work by Johnson³ showed typical oscilloscope patterns for the various modes of electrical behavior, nor did the HYGAS observers' trace indicate gross arcing above the surface (Figure 4a.1-5). Researchers working with the 4-inch unit therefore concluded that their region of operation was out of the Ohm's law region and in the "intermediate" region. In the velocity range studied, the resistivity was proportional to the bed expansion ratio.

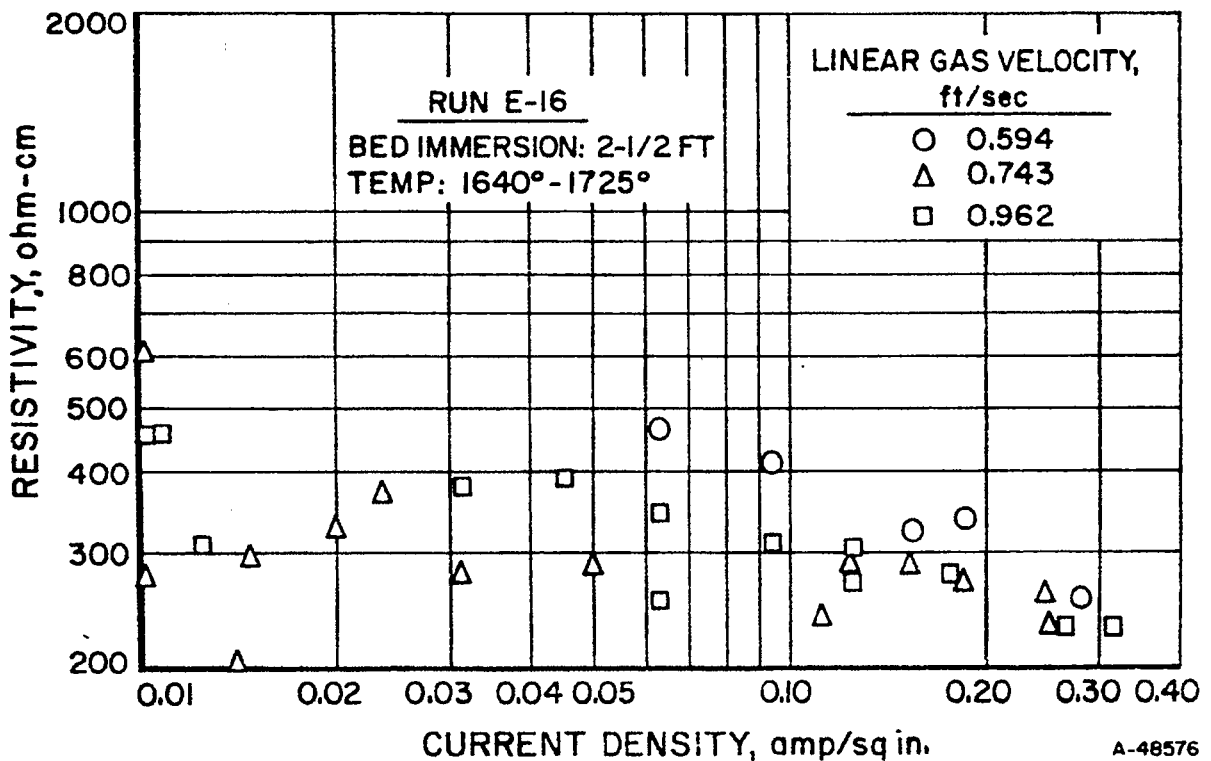


Figure 4a.1-1. EFFECT OF CURRENT DENSITY ON RESISTIVITY AT 40 lb/sq. in.

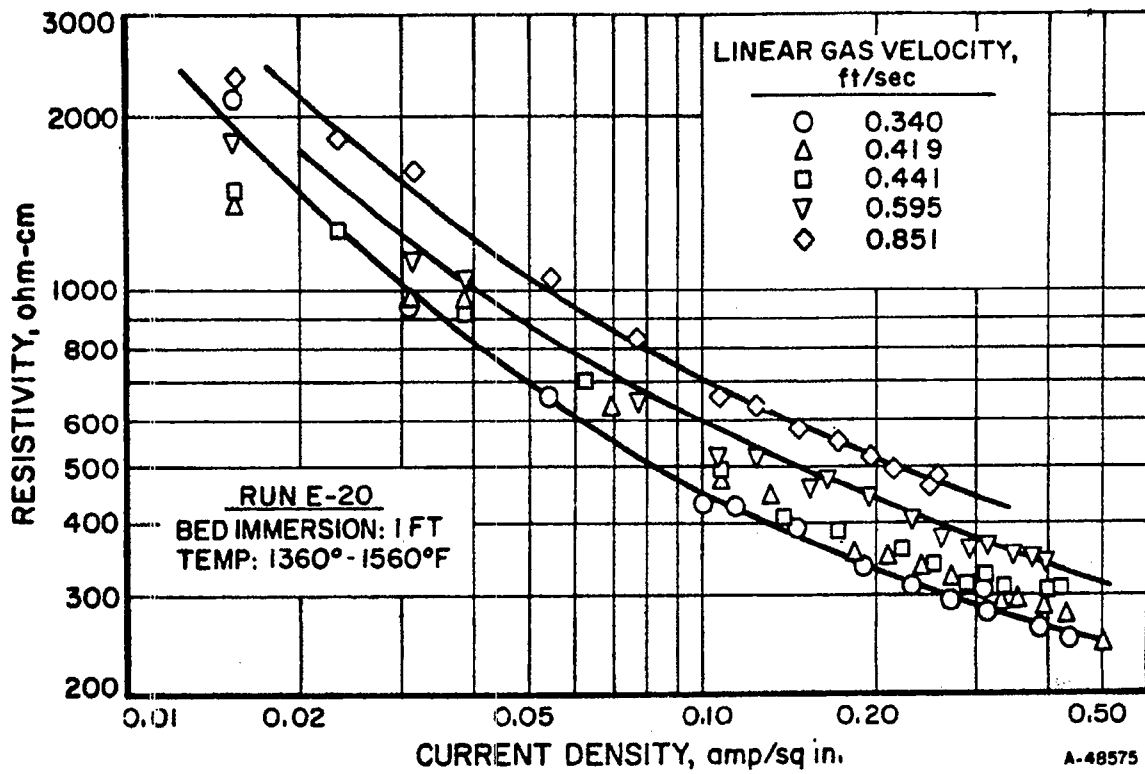


Figure 4a.1-2. EFFECT OF CURRENT DENSITY ON RESISTIVITY AT 210 lb/sq. in.

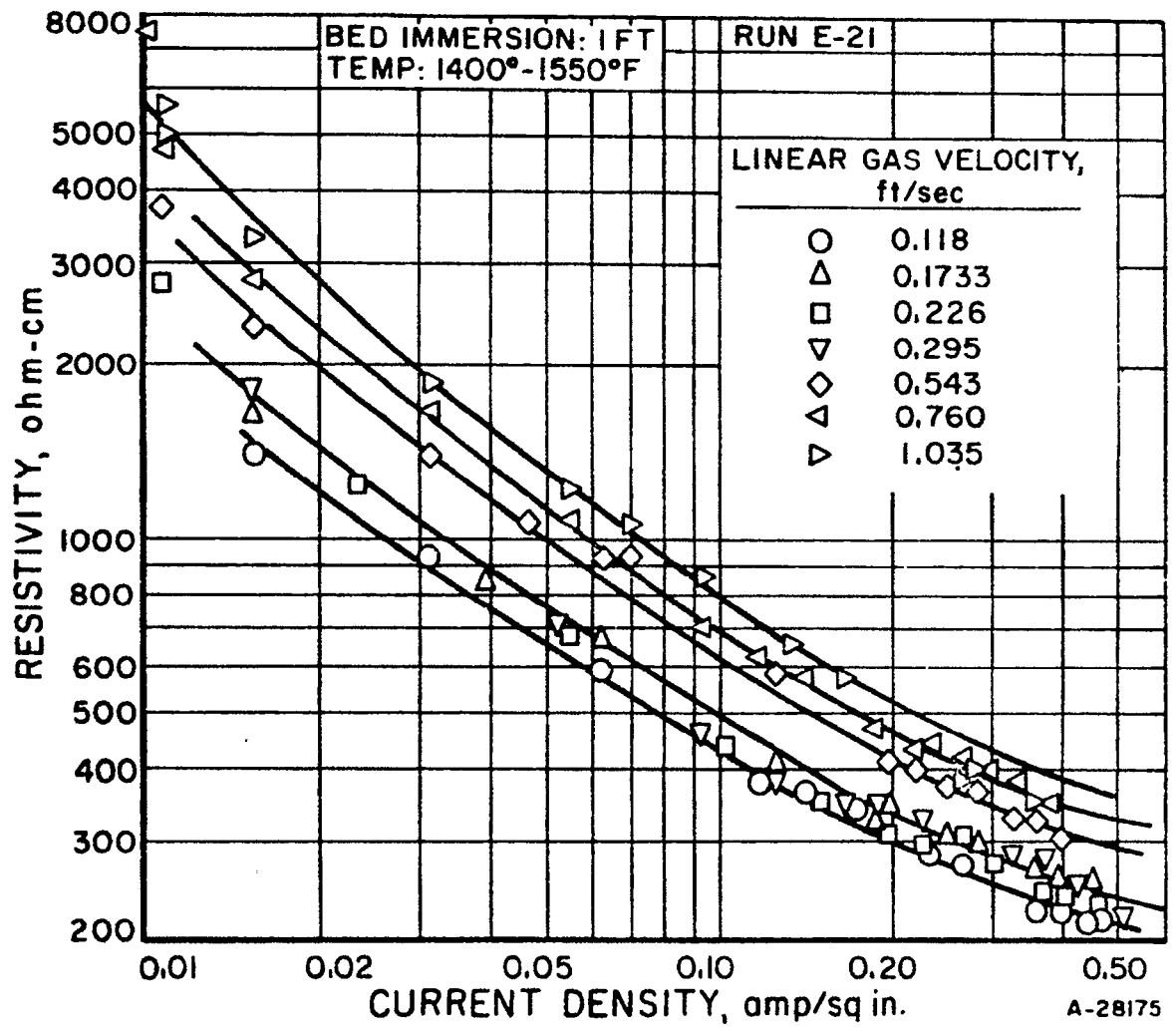


Figure 4a.1-3. EFFECT OF CURRENT DENSITY ON RESISTIVITY AT 530 lb/sq. in.

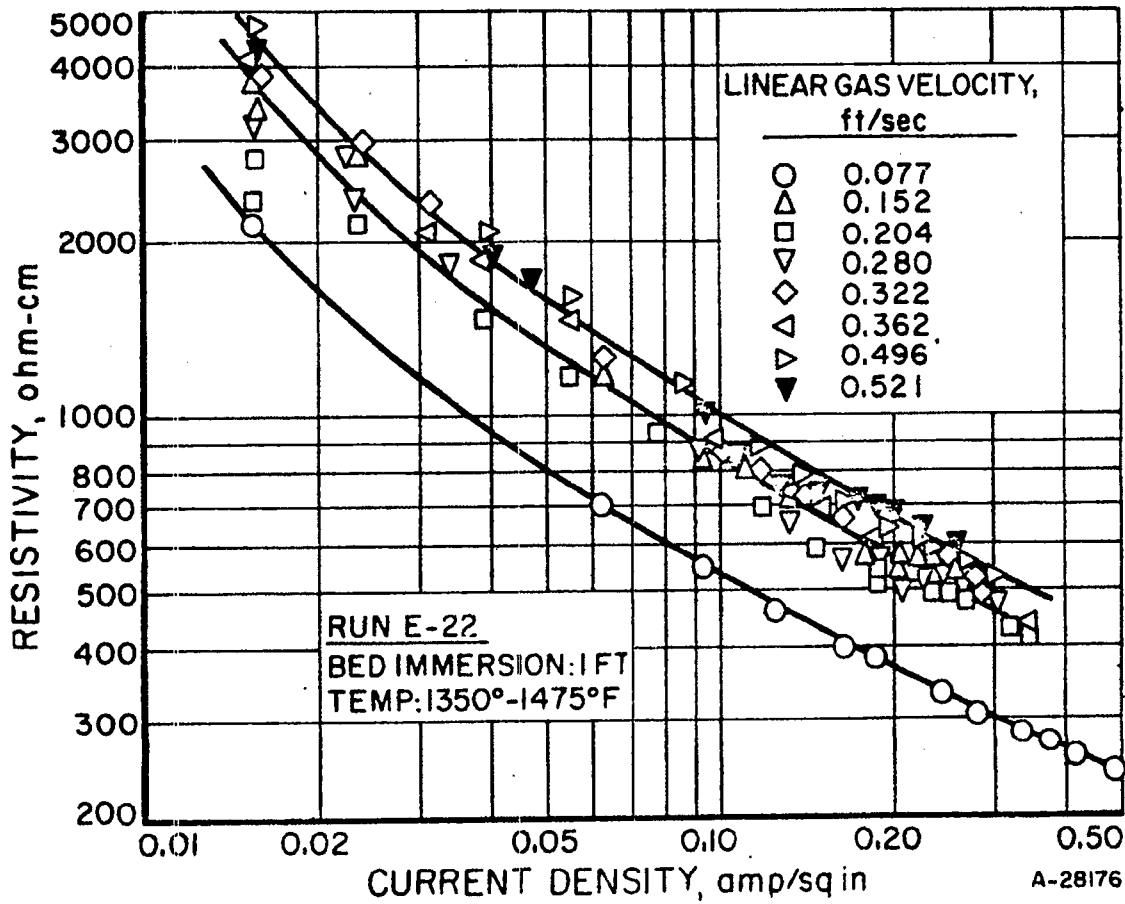
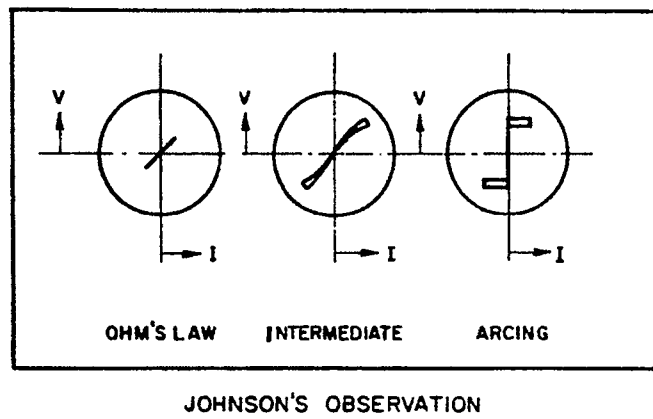
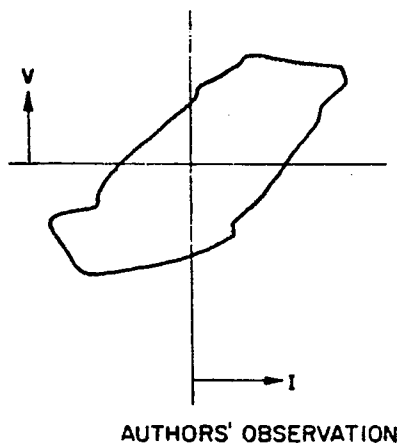


Figure 4a.1-4. EFFECT OF CURRENT DENSITY ON RESISTIVITY AT 1040 lb/sq. in.



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Figure 4a. 1-5. OSCILLOSCOPE TRACE OF VOLTAGE-CURRENT PATTERN IN FLUID BED

The mechanism for current flow in a high-pressure fluidized bed is through spark discharge between bed particles across gas gaps whose average magnitude is characterized by the bed expansion ratio. Current flow is analogous to sparking discharge between spherical electrodes in a gaseous atmosphere. The discharge follows a modified Paschen's law, whereby the resistivity is proportional to pressure at low values of current density, but the dependence on pressure decreases with increasing current density until, at 0.5 amperes per square inch, the resistivity is essentially independent of pressure as illustrated in Figure 4a.1-6.

4a.1.2 Equipment

A process flow and instrument diagram is illustrated in Figure 4a.1-7. Reactants in the continuous system flow countercurrently. The spent hydrogasified char was screw-fed from a balanced-pressure hopper into the reactor top; process steam entered through a distributing nozzle at the reactor bottom. Reacted char was screw-discharged from the reactor bottom to a residue receiver vessel. Both feed and discharge screws were hydraulically operated and equipped with panel-mounted variable-speed controls. The make-gas exited at the reactor top into a condenser-cooler and mist filter. A liquid-collecting tank was located below the condenser for storage of unreacted water. After pressure letdown, the gas was metered and flared to the atmosphere. For operation at high reactant throughput rates in the 300 pounds per hour range, the condenser section was modified by installing a water-quench tower.

The reactor vessel (Figure 4a.1-8) was 9 feet long and consisted of an outer pressure shell of 24-inch Schedule 80 carbon steel pipe in which a 6-inch Schedule 5 Type-316 stainless steel pipe was inserted. The annulus between the two was compacted with bulk-fiber ceramic insulation. The inner pipe was joined to the bottom reactor hub by a stainless steel connecting sleeve and acted as one electrode in the system. The top closure on the reactor was filled with castable firebrick insulation with openings to allow for char feeding, make-gas exiting, electrode entering, and pressure monitoring. The center electrode entered the reactor through a water-jacketed section at the top of the closure and was electrically insulated from the reactor. A nitrogen purge entered just below the electrode entrance of the water-jacketed section to prevent char particles from collecting in the annulus. A Teflon sleeve shielded the electrode the length of the cooling section.

The power source was a 450-horsepower, 2300-volt, 3-phase, 60-cycle, 100-ampere a-c motor. The motor was directly coupled to a 300-kilowatt d-c continuous capacity and up to a 600-kilowatt short-time output. A silicon control rectifier (SCR) system allowed continuous voltage control between 50 and 550 volts, and current control between 200 and 1600 amperes. The circuits were fused with fast-acting fuses.

Both voltage- and current-limiting controls function by adjusting the field excitation of the generator. The output voltage was measured by the voltage drop across the interpole. After observers consistently noted the absence of current control, however, an external resistor stack was installed and the voltage drop across it was used to indicate current. This arrangement operated successfully. HYGAS researchers determined that, with the highly fluctuating current in the system, the iron core of the interpole had generated self-induced voltages that shielded the IR drop across the interpole.

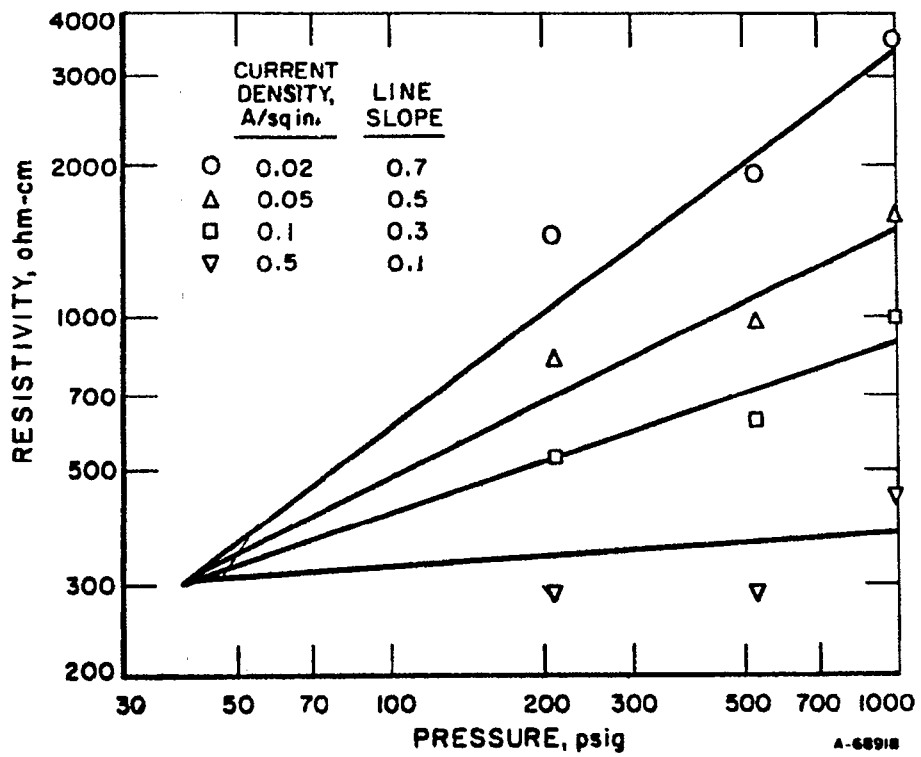


Figure 4a.1-6. EFFECT OF PRESSURE ON RESISTIVITY AT 0.5 ft/s

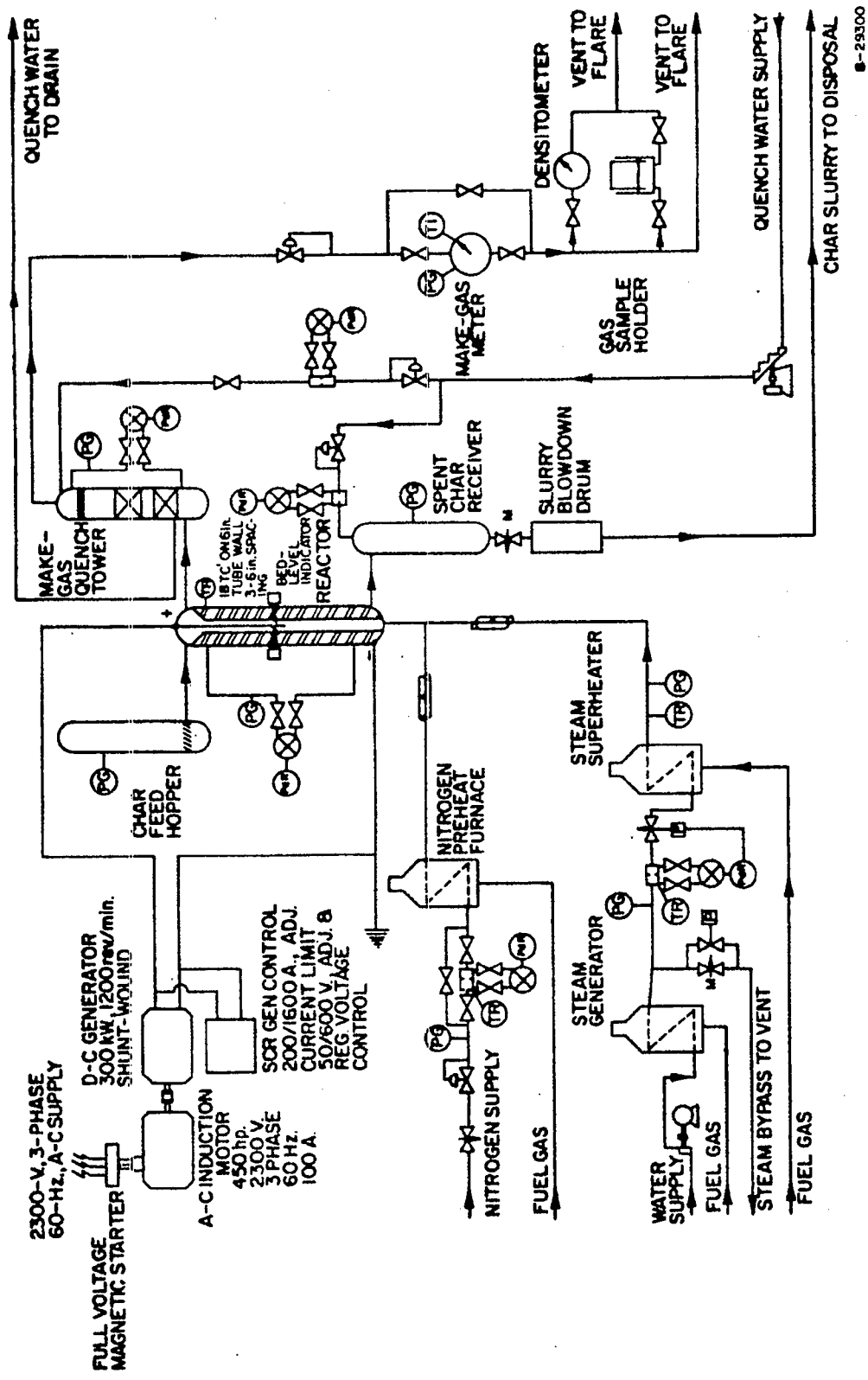


Figure 4a.1-7. PROCESS FLOW AND INSTRUMENT DIAGRAM

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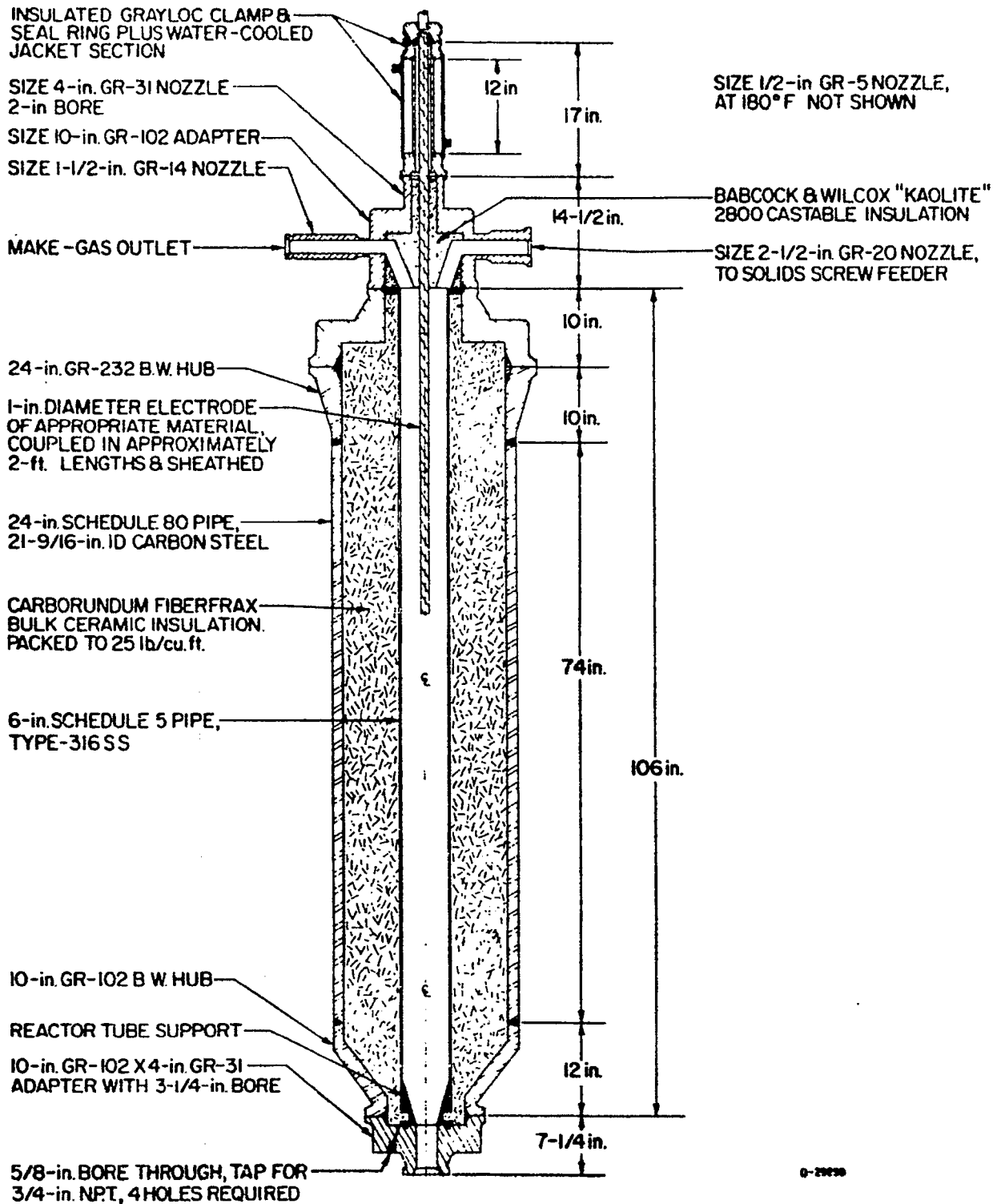


Figure 4a.1-8. CUTAWAY VIEW OF REACTOR VESSEL

Nitrogen, used as the fluidizing gas during the reactor heat-up period, was monitored by an orifice meter and preheated in a gas-fired furnace before entering the reactor.

Steam was supplied to the reactor by a gas-fired high-pressure steam generator rated at a continuous 300 pounds per hour flow in line with a high-pressure steam superheater capable of producing 1250° F steam at 1250 pounds per square inch (gage). The steam flow rate was controlled and measured by an orifice-metering system located in the transfer line between the steam generator and superheater. Because of the constant output of the steam generator, a venting system upstream of the orifice provided for flow control at variable rates below the 300 pounds per hour rating of the generator.

Bed-temperature measurements were provided by 18 thermocouples attached to the outer wall of the 6-inch tube, spaced at 3- to 12-inch intervals along the length of the tube. The thermocouple leads exited through pressure glands in the reactor bottom closure. Temperatures were recorded at the control panel.

The char-bed level was indicated by the pressure differential across the reactor and with a radiation density gage mounted on the outer reactor shell. The radiation gage included a gamma-ray source that emitted a beam through the reactor, at a predetermined level, in line with a detector on the opposite wall. The detector output was inversely proportional to the density in the 6-inch reactor, and was relayed to a panel-mounted indicator.

4a.1.3 Experimental Procedures

After a satisfactory pressure test, the feed hopper was loaded with enough hydrogasified char to run through an 8-hour shift. The system was pressurized to run conditions with nitrogen, and a nitrogen flow rate was established to ensure fluidization of the bed before power was applied. About 10% of the total nitrogen flow was used to purge the reactor top. Char was screw-fed into the reactor until the desired bed level was attained, as indicated by the radiation detector and reactor differential pressure. The power was turned on to an initial power input of 1 to 2 kilowatts while the stability of the system was checked. The power was then gradually increased to from 10 to 15 kilowatts. Bed temperatures of 1800° F were reached within 2 to 3 hours. Figure 4a.1-9 illustrates the typical bed temperature-power input relationship, and Figure 4a.1-10 illustrates the overall resistance behavior during a typical heat-up period. These compare generally with results reported by Goldberger *et. al.*⁴ and Iowa State University.⁵ The nitrogen superficial velocity through the reactor at the start of the heating period was slightly higher than that required for incipient fluidization, and was held constant during the heat-up period, eventually reaching several times the incipient fluidization velocity.

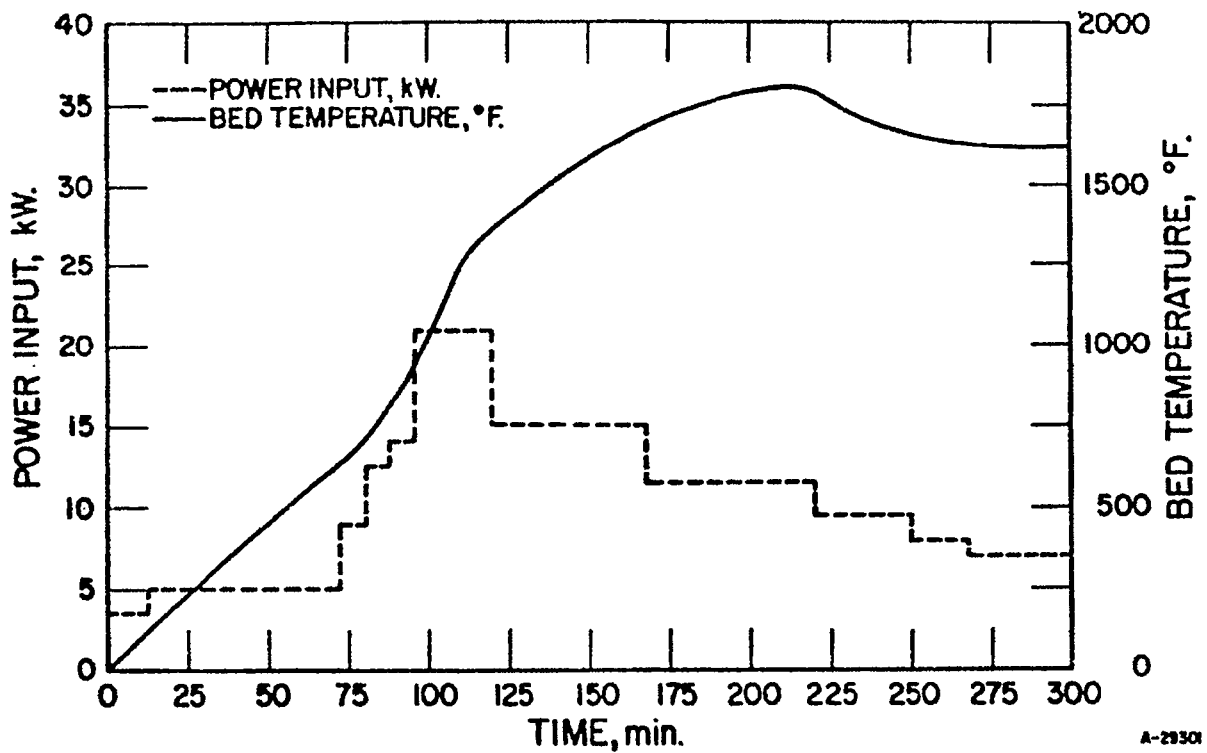


Figure 4a.1-9. POWER-TEMPERATURE RELATIONSHIP OF NITROGEN AS FLUIDIZING GAS AT BED HEIGHT OF 5 ft AND INNER ELECTRODE IMMERSION OF 24 in.

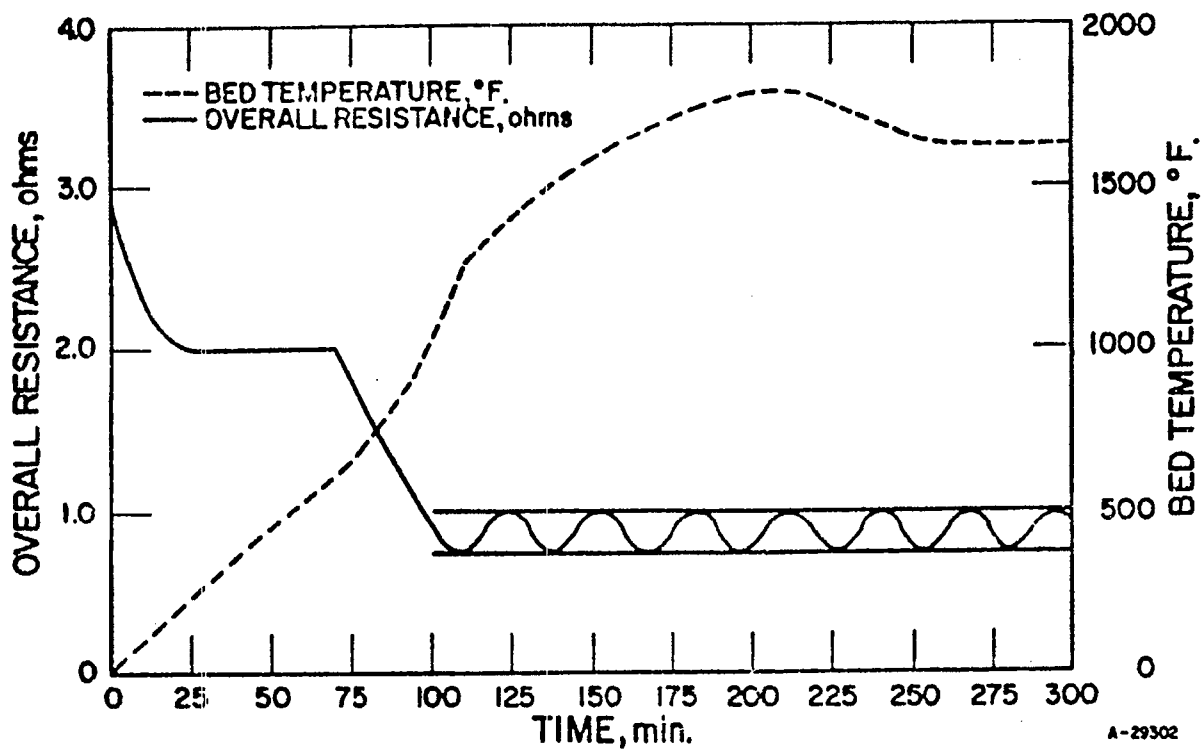


Figure 4a.1-10. OVERALL RESISTANCE-BED TEMPERATURE RELATIONSHIP DURING REACTOR HEAT-UP

As the bed temperature approached run conditions, the steam generator was started and the steam was vented to the atmosphere. Before switching to steam, the power input was lowered to approximately 1 kilowatt. The process steam was directed into the reactor, and the nitrogen flow rate was simultaneously decreased until just the nitrogen purge and steam entered the reactor. The power input was then increased to maintain the desired bed temperature. The char-feed screw was operated at a rate calibrated for a particular char-feed input. The discharge screw was adjusted to the rate necessary to maintain the bed level indicated by the bed-level detector.

During steady-state operating periods, the make-gases were continually metered, sampled for mass spectrometry analyses, monitored by a densitometer, and flared to the atmosphere. Unreacted water was periodically drained from a collecting tank below the condenser-cooler. Spent char samples were collected from the residue following the completion of the test, weighed for material balances, and analyzed.

4a.1.4 Shakedown and Start-Up Problems

The uniqueness of the electrothermal gasification concept, particularly at high pressures, led to a substantial effort in establishing safe, reliable system operating procedures during the program. The following discussion briefly relates a number of the operating problems encountered, and the steps that were taken to overcome them.

Initially, the pressure-sealing closure through which the center electrode entered the reactor was electrically isolated from the reactor by a non-conducting coating of molybdenum disulfide and teflon on the pressure seal ring. The ring was enclosed by a holding clamp coated with non-conducting polyurethane. The connection was rated at 20 megohms resistance, 1000-volts direct current, 1000 pounds per square inch (gage) and 200°F, as illustrated in Figure 4a.1-11.

This setup worked well at operating pressures up to 500 pounds per square inch, but at 1000 pounds per square inch the coating on the pressure-seal ring would "cold flow", causing a short circuit across flange faces and an arcing condition which prevented operation of the unit. This problem was overcome by replacing the insulated metal pressure closure with a standard high-pressure bolted flange arrangement. A teflon pressure gasket between flange faces and insulated flange bolts provided the electrical isolation between the high potential center electrode and the low potential reactor structure, as shown in Figure 4a.1-12. No further short-circuiting problems were encountered in this area.

The char bed level was initially determined by monitoring the pressure differential across the reactor. Continual plugging of the pressure taps from steam condensation and char fines deposits caused the differential-pressure transmitter to malfunction. The installation of the bed-level radiation detector provided a reliable means to obtain these vital measurements.

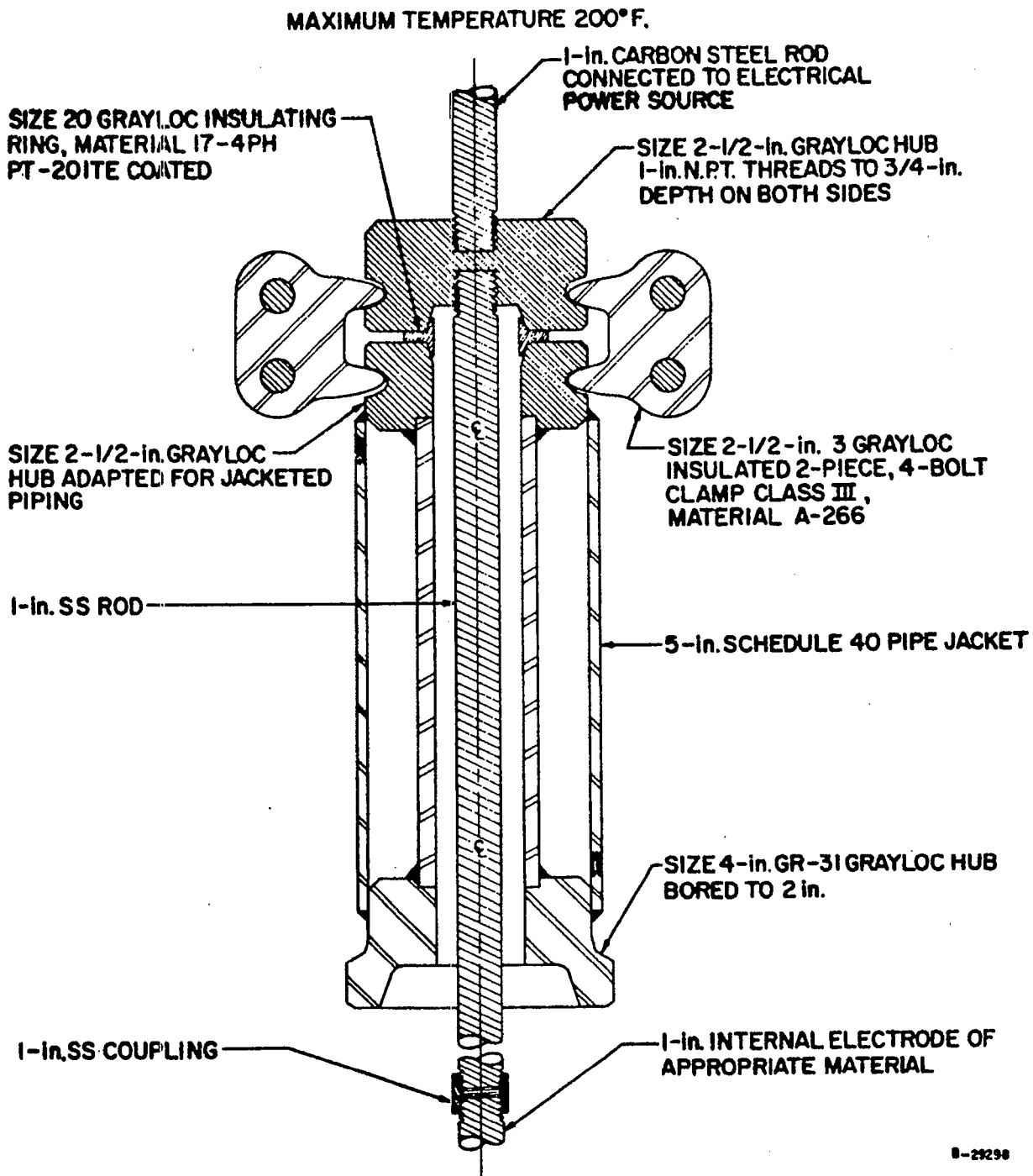


Figure 4a.1-11. INITIAL PRESSURE CLOSURE FOR CENTER ELECTRODE ENTERING REACTOR

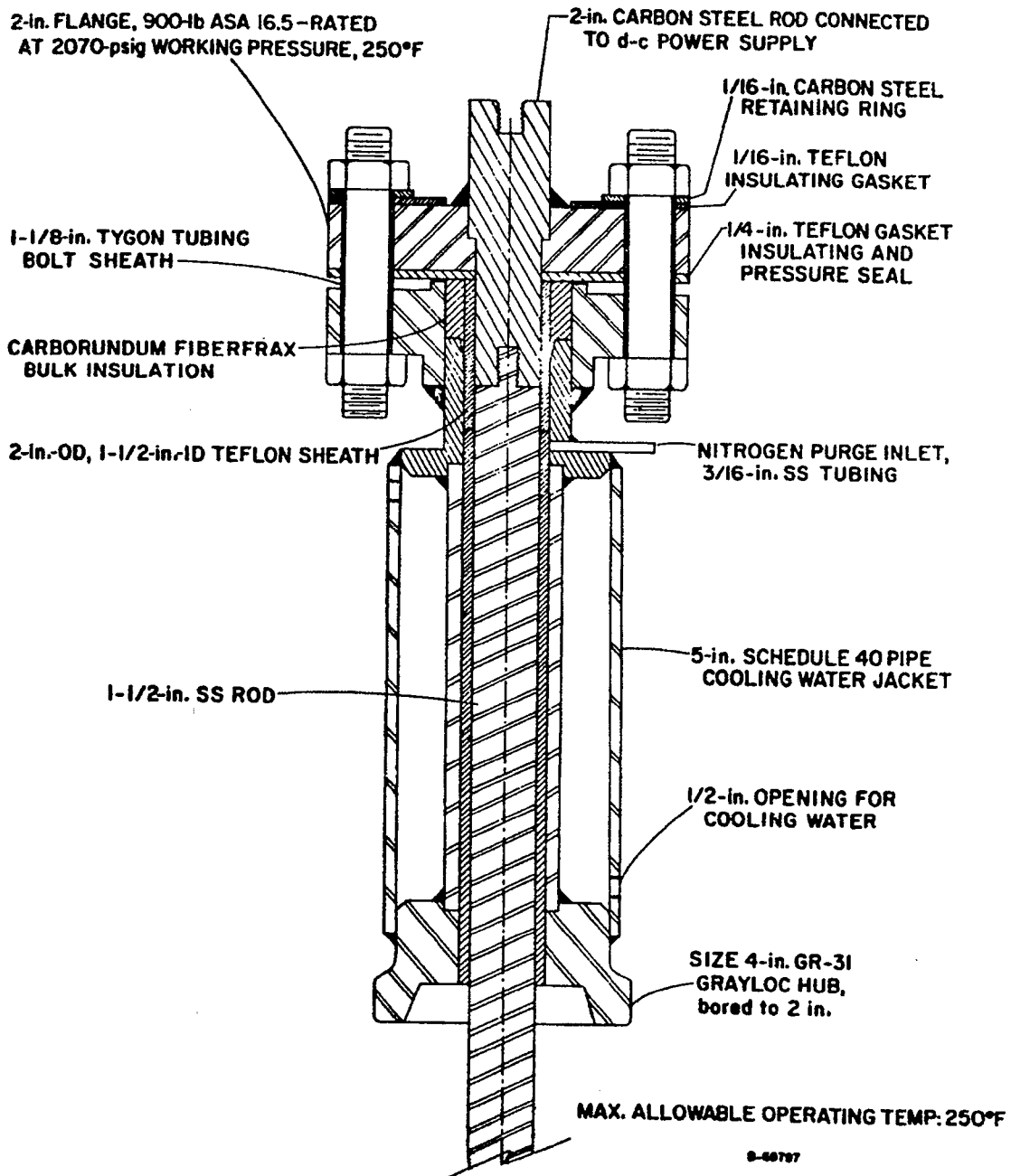


Figure 4a.1-12. INSULATED, FLANGED PRESSURE CLOSURE
AT CENTER ELECTRODE ENTRANCE INTO REACTOR

The make-gas transfer line to the condenser-cooler and the char feed inlet occasionally plugged with steam condensate and char fines, but resistance heaters placed on the transfer line and feed screw housing eliminated the problem.

Pressure imbalances among the vessels caused unloading of solids from the feed hopper into the reactor or from the reactor into the char residue receiver. The problem was solved by installing pressure-equalizing lines between the vessels, and a nitrogen purge at the top of the reactor.

The most critical point in the operation of the unit occurred during the transition from fluidizing nitrogen to process steam. The erratic gas flow rates and pressure upsets that occurred were similar to those experienced in the calcination of fluid coke, when green coke is fed into the reactor after reaching run temperature.⁶

The increased flow of off-gases resulting from the steam-carbon reaction caused slugging in the bed, excessive solids loading in the gas exit, and subsequent plugging of the line. The pressure drop caused by the plugging suppressed the fluidization of the char bed, caused short-circuiting through the bed, melting of the center electrode tip, and slagging of the char. The melting of the electrode tip and the damage to the reactor wall often occurred before the fast-acting fuses could open the circuit. It was therefore imperative that proper fluidization be maintained while the power was being applied to the reactor.

The unstable condition which occurred during the transition from fluidizing nitrogen to steam was kept to a minimum by decreasing the power input, while simultaneously increasing the steam flow rate and decreasing the nitrogen flow rate. The transition was made at a lower bed temperature, thus at a lower off-gas rate; the system was then brought to the desired run conditions.

Among mechanical problems encountered were the malfunction of the voltage control in the SCR controller of the d-c generator, and the failure of the steam superheater, caused by a faulty safety interlock which led to the rupturing of the coil.

4a.1.5 Electrode Configurations

4a.1.5.1 Concentric Electrodes

The concentric electrode configuration consisted of an electrode inserted through the reactor top and immersed in the fluidized char bed. The inner reactor liner was of a conducting material and acted as one electrode in the system. The overall resistance of the system could be varied by changing the immersion depth of the electrode or the reactor diameter and thus the length of current path. During the initial shakedown and operation of the unit, the concentric electrode configuration, with a 1.5-inch-diameter stainless steel electrode at a 2-foot electrode immersion into the bed, provided the smoothest operation; with some variations, this configuration was used in all successful gasification tests.

4.1.5.2 End-to-End Electrodes

In the end-to-end electrode configuration (Figure 4a.1-13), the current traveled from an electrode inserted through the reactor top and immersed in the char bed, and along the length of the bed, to ground at the bottom of the reactor. The inner reactor wall consisted of a nonconducting material: either mullite, a lightweight refractory cement, or vycor. Of particular interest were:

- The effect of electrode spacing on the overall resistance of the system, and
- The effect on electrode life caused by the higher current density that resulted from the decreased area of current flow.

An incentive for operating end-to-end electrodes was the expected high voltage-low current operation that would minimize I^2R losses for a given power input.

A series of tests was conducted at electrode spacings of 1 to 12 inches using a 1.5-inch stainless steel rod as the top and bottom electrodes. In efforts to increase the area of current flow, several variations in the bottom electrode set-up were tested, but without success. The overall resistance during the runs ranged from 50 to more than 300 ohms. Because the d-c generator was limited to a 550-volt output, it was not possible to obtain the necessary power to conduct a high-temperature test. Further study was concentrated on the concentric electrode arrangement.

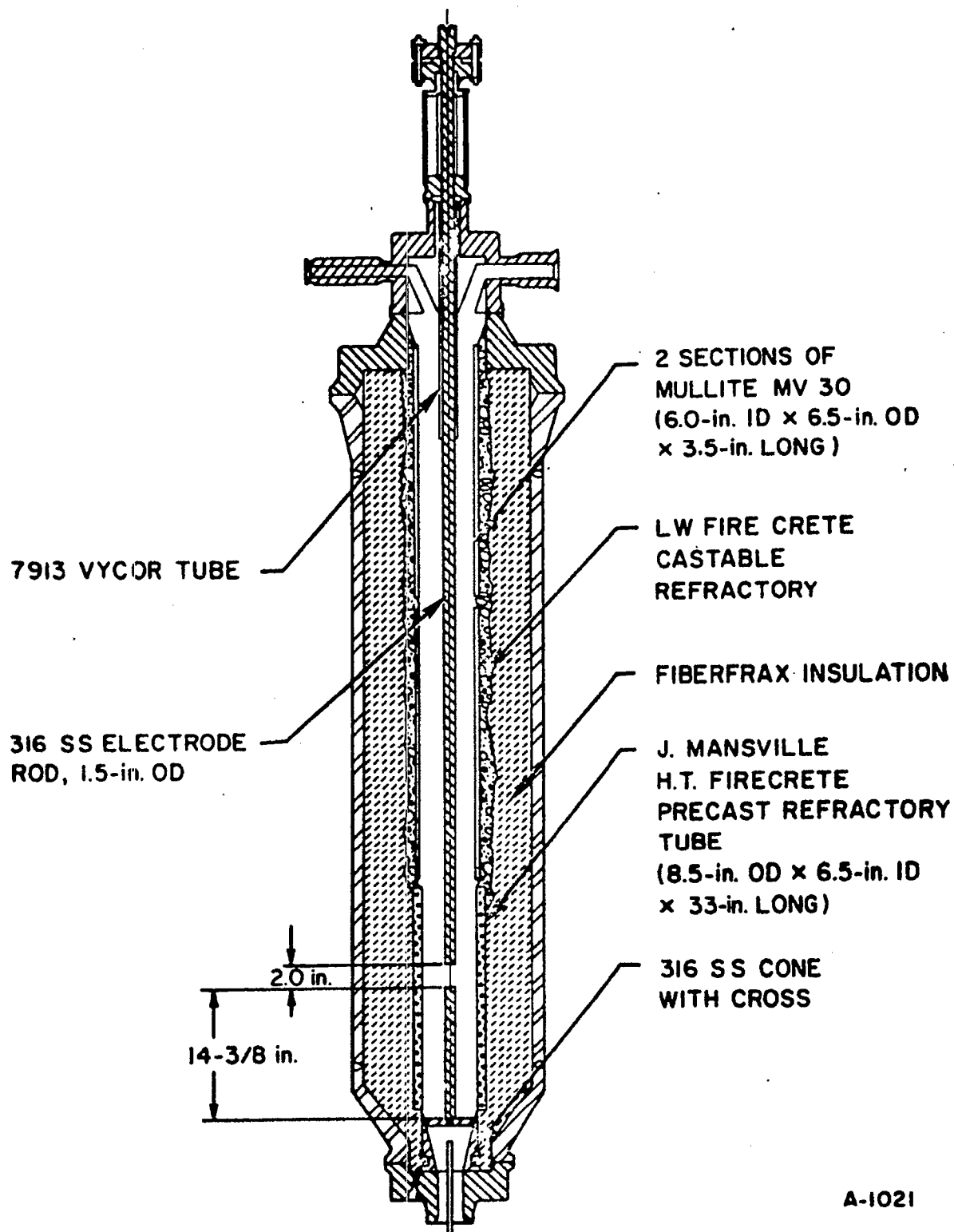
4a.1.5.3 Multiple Electrodes

One test was conducted to observe the operational characteristics of a multiple-center electrode configuration (Figure 4a.1-14). The bottom 36 inches of the center electrode consisted of three 0.625-inch Type 316 stainless steel rods welded at 120-degree intervals to a 1.5-inch Type 316 stainless steel electrode. During the reactor heat-up, a leak developed at the base of the reactor and observation of the electrical characteristics of the configuration proved inconclusive.

4a.1.6 Electrode Materials

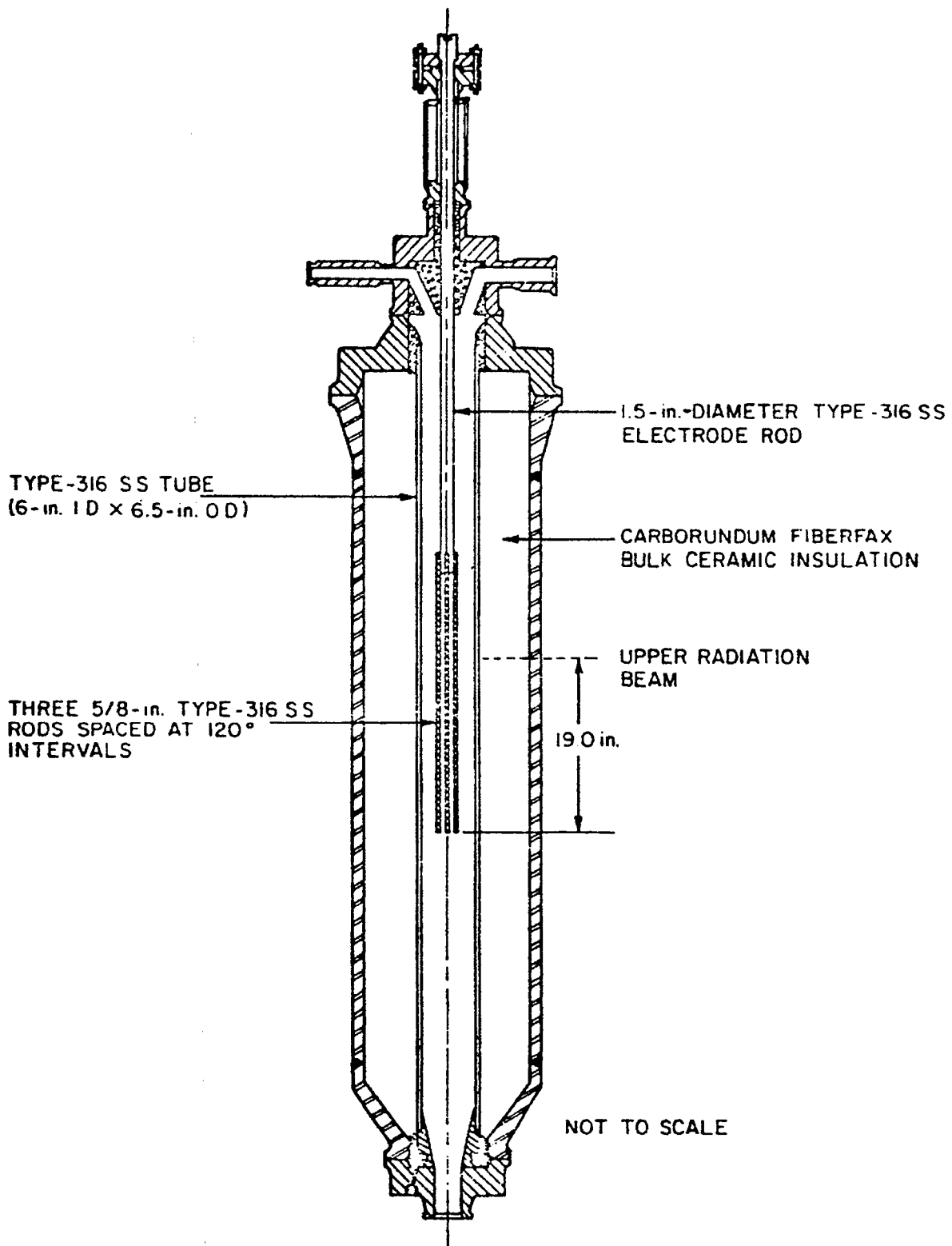
4a.1.6.1 Metallic

Successful tests were conducted with types 316, 430 and 440 stainless steels as both the center (1.5-inch) and outer (6-inch tube) electrodes. In addition, Type 17-4 PH (Armco Steel) and molybdenum were tested only for the center electrode. All the metals tested exhibited satisfactory electrical properties for operation. After one test, however, the molybdenum electrode became severely blistered and coated with a heavy ash-like material of higher electrical resistance (5 meg-ohms) which prevented its use in further testing.



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Figure 4a.1-13. CUTAWAY VIEW OF REACTOR VESSEL
END-to-END CONFIGURATION



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Figure 4a 1-14 CUTAWAY VIEW OF REACTOR VESSEL
WITH MULTIPLE CENTER ELECTRODES

The stainless steels functioned well under gasification conditions and became only slightly pitted during normal operation. A very thin semi-conductive oxide coating would form on stainless electrodes during the cooling of the reactor, but required only a light brushing of the surface between tests to lower the resistance. The availability of the Type 316 stainless steel led to its use in most of the runs as both center and outer electrodes. Several Type 316 stainless steel rods operated successfully in excess of 100 hours each.

4a.1.6.2 Silicon Carbide

Bonded silicon carbide has excellent physical and chemical properties in a high-temperature, high-pressure reducing atmosphere. These properties led to the consideration of bonded silicon carbide as a possible electrode material in electrothermal gasification. The compound is impervious to attack by high-pressure steam and hydrogen. It exhibits excellent refractory properties and thus is not affected by extreme temperatures. Finally, it is electrically conductive at the operating temperature of the electrothermal reactor.

A series of tests were conducted with a silicon carbide center electrode and a stainless steel outer electrode, and the reverse, a silicon carbide outer electrode and a stainless steel center electrode. Although operating temperatures of 1800⁺°F were obtained, the tests were terminated prematurely because the silicon carbide fractured in either application.

This is attributed to the brittleness and low tensile strength of silicon carbide.

4a.1.7 Chars Tested

Successful tests were conducted with high-volatile bituminous chars, medium-volatile bituminous char, and with lignite char, all produced in the 4-inch hydrogasification test unit. The inventory of hydrogasified char was insufficient for the electrothermal program and most of the tests were conducted with a char from the FMC Corporation Project COED pilot program. The characteristics of this char closely resembled bituminous char residue from the 4-inch hydrogasifier.

For comparison of the various chars, the proximate and ultimate analyses of typical feed material are presented in Table 4a.1-1. In all tests, the feed was dried and screened to a nominal -10+80 mesh U.S. sieve size.

4a.1.8 Gasification Results

The primary purpose of the electrothermal gasification program was to specify conditions which would be of value to engineers in the design of an electrothermal gasification system for integration into the HYGAS pilot plant. The scope of the testing in the 6-inch unit was, therefore, confined to the definition of criteria which would satisfy heat and material balances and synthesis gas yields required at the planned operating conditions of that plant. The tests at pressures below 1000 pounds per square inch (gage) were conducted mainly for the operational shakedown of the unit.

Table 4a. 1-1. CHARACTERISTICS OF VARIOUS FEED CHARs

| | EG-12 | EG-14 | EG-78 | EG-75 |
|---------------------------------|--|--------------------------|---|--|
| | Hydrogasified Ireland Mine <u>Bituminous</u> | FMC <u>Bituminous</u> | Hydrogasified <u>Montana Lignite</u> | Hydrogasified <u>North Dakota Lignite</u> |
| Proximate Analysis, wt % | | | | |
| Moisture | 1.5 | 2.2 | 3.8 | 1.8 |
| Volatile Matter | 4.3 | 5.0 | 7.3 | 7.7 |
| Ash | 17.2 | 17.5 | 15.5 | 13.8 |
| Fixed Carbon | <u>77.0</u> | <u>75.3</u> | <u>73.4</u> | <u>76.7</u> |
| Total | 100.0 | 100.0 | 100.0 | 100.0 |
| Ultimate Analysis, wt % | | | | |
| Ash (dry) | 17.42 | 17.91 | 16.07 | 14.04 |
| Carbon | 78.70 | 76.00 | 77.90 | 77.00 |
| Hydrogen | 0.99 | 0.92 | 1.23 | 1.18 |
| Sulfur | 1.65 | 2.22 | 0.37 | 0.80 |
| Oxygen | 0.63 | 1.81 | 4.02 | 4.49 |
| Nitrogen | <u>0.61</u> | <u>1.14</u> | <u>0.41</u> | <u>0.49</u> |
| Total | 100.00 | 100.00 | 100.00 | 100.00 |

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Operating conditions and results of all successful tests and the analyses of the feed and residue chars are tabulated in Appendix 4a.1-A.

The principal reactions of the steam gasification of coal char at high temperature and pressure are -



4a.1.8.1 Steam-Carbon Reaction

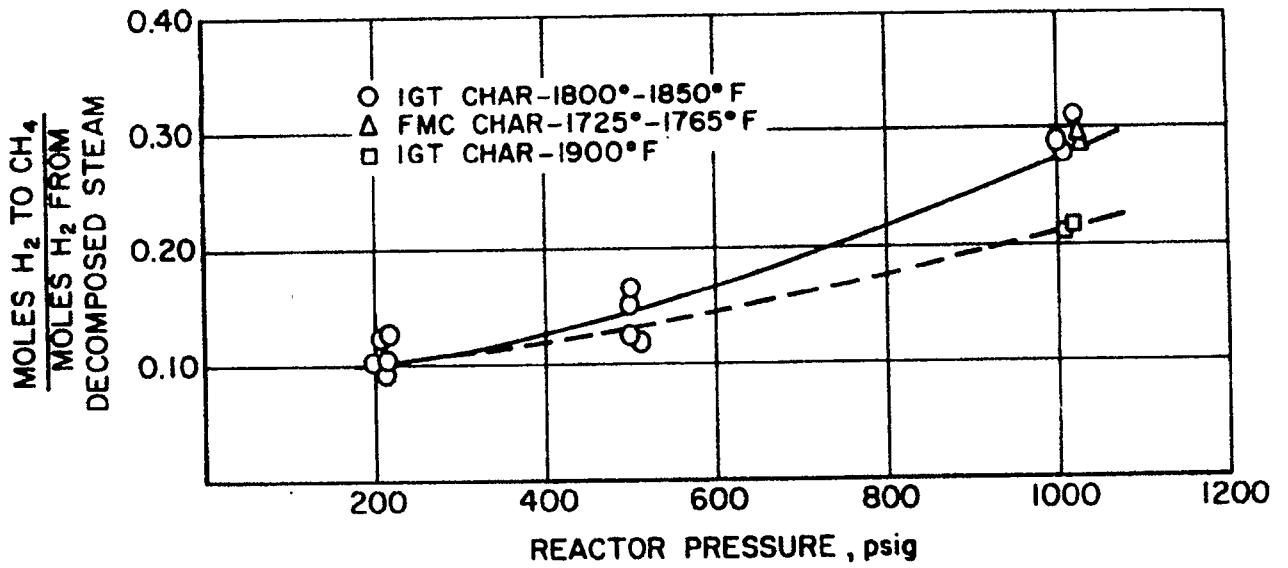
The reaction rate of the highly endothermic and temperature-dependent steam-carbon reaction (1) is quite low at temperatures less than 1700° F.¹⁰ Because substantial carbon gasification was desired, the operating range of temperatures was generally above 1800° F. In several tests conducted at 500 psig the steam-carbon reaction was given residence times exceeding 30 minutes in order to determine whether the carbon gasification and synthesis-gas yields were equilibrium-limited; they were not. In tests at 1000 pounds per square inch (gage) with bituminous char as feed, temperatures of 1850° to 1900° F, char residence times of 10 minutes, and steam/char feed ratios of 1.5 pounds per pound were required to produce the desired carbon gasification and synthesis-gas yields. At the specified conditions, the product-gas concentrations correspond to an equilibrium at temperatures 200° to 300° F lower than the bed temperature. Tests with hydrogasified lignite char as feed also produced the required synthesis gas yields at similar residence times and steam/char feed ratios, but at much lower average bed temperatures (~1650° F). The rapid reaction rate, as evidenced by the short char residence time with any char, later tended to hold down the size of the reactor required in the design and construction of the HYGAS pilot plant, and will have the same influence on larger plants utilizing the HYGAS process.

4a.1.8.2 Water-Gas Shift Reaction

The rate of this reaction (2) is quite rapid at the temperatures discussed; the reaction reached equilibrium in nearly all of the runs conducted.

4a.1.8.3 Methane Formation

The methane formation reaction (3) also reached equilibrium in most cases. The methane yield in the product gas increased, as expected, at higher pressures. In tests at 1000 pounds per square inch (gage) and temperatures of 1800° to 1850° F, the ratio of hydrogen going to methane, to the total hydrogen yield from decomposed steam, was 0.3 mole/mole. In order to allow maximum methane formation and hence heat release in the hydrogasifier, it became preferable to hold down the methane content in the gas from the electrothermal gasifier; this ratio has been reduced to 0.2 mole/mole by operating at temperatures near 1900° F, as illustrated in Figure 4a.1-15.



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Figure 4a.1-15. RELATION OF MOLES H₂ TO CH/TOTAL H₂ FROM DECOMPOSED STEAM TO REACTOR PRESSURE

4a.1.8.4 Carbon Gasified

Both reactor temperature and the steam-to-char feed ratio have pronounced effects on the carbon gasification rate. At 1800 to 1900° F a steam-to-char feed ratio of 0.8 to 1.5 pound per pound, 40 percent or more of the carbon can be gasified in about 10 minutes using hydrogasified bituminous chars.

In most instances, the FMC bituminous char showed somewhat higher carbon gasification under similar conditions. The hydrogasified lignite chars, however, required a bed temperature of less than 1700° F to obtain 50% carbon gasification at steam/char feed ratios of 0.8/1.5 pound per pound, and at residence times of 10 to 15 minutes.

4a.1.8.5 Synthesis-Gas Yields

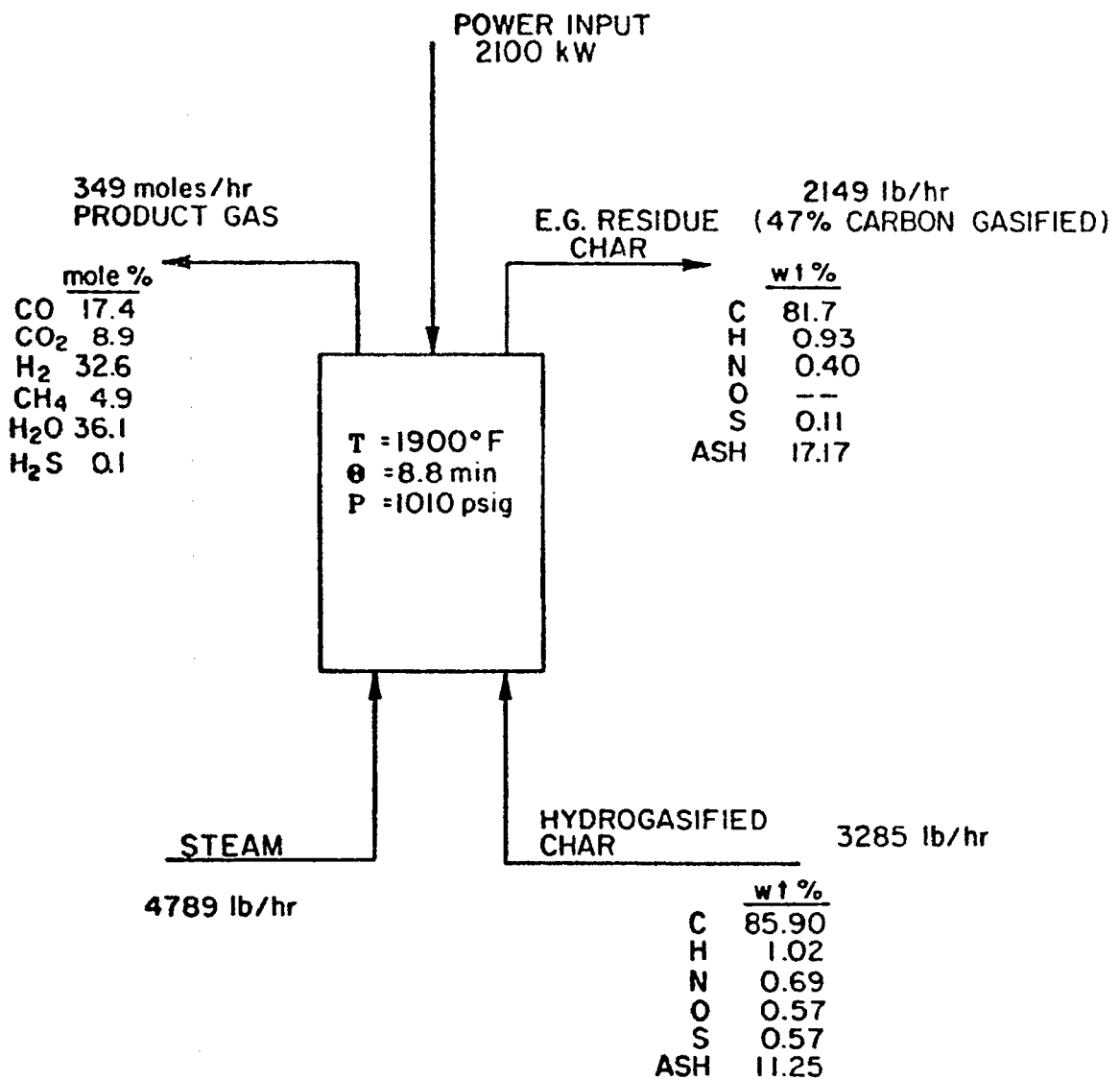
In the tests conducted at 1000 pounds per square inch (gage), the yield of equivalent hydrogen — i. e., hydrogen plus carbon monoxide in the product gas — ranged from 12 to 20 standard cubic feet per pound of char fed at the various run conditions.

Once the initial test conditions of the HYGAS plant had been set, the electrothermal gasification study was directed toward obtaining synthesis-gas yields that would supply the required feed gas to the HYGAS reactor while utilizing the expected char discharge rate from the reactor. A material balance scaled from data obtained in test E. G. -48 (Figure 4a.1-16), reflects the requirements of the hydrogasification section of the HYGAS plant at run conditions calling for the predicted maximum synthesis gas feed. As can be seen, a bed temperature of 1900° F, char residence time of 10 minutes and a steam-to-char feed ratio of 1.46 pound per pound resulted in a hydrogen plus carbon monoxide yield of 20 standard cubic feet per pound of char fed. These results satisfy the HYGAS requirements. A power input of 67 kilowatts was applied during the test, including heat losses, which extrapolates to 2100 kilowatts at the scale of the HYGAS plant feeding 3 tons per hour of bituminous coal. This synthesis gas requirement is the highest anticipated for either bituminous or lignite chars in the HYGAS plant.

4a.1.9 Electrical Characteristics

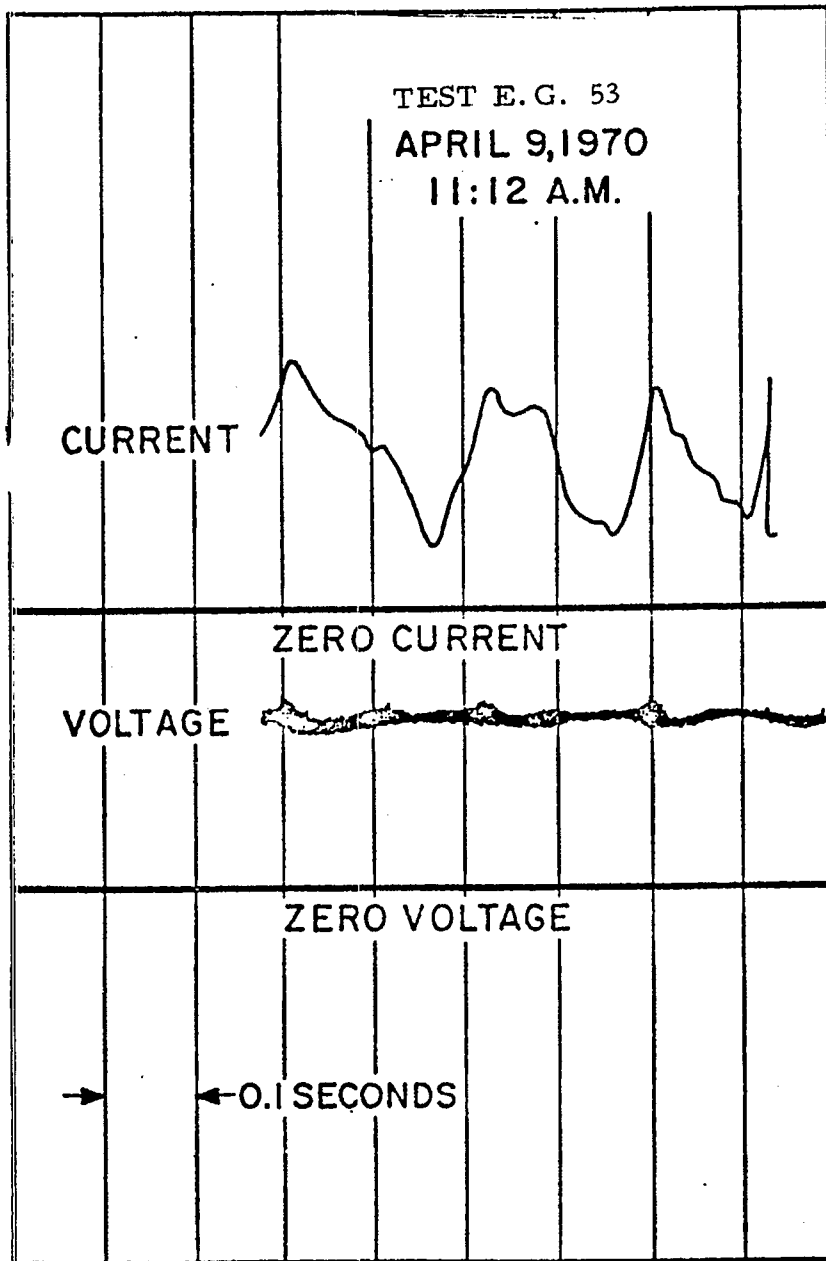
4a.1.9.1 Voltage-Current Relationship

As was discussed earlier, the electrothermal gasifier is a nonlinear resistive load; because of bed fluctuations, the load is unstable, varying in resistance over wide ranges and thus — in the case of a constant voltage source — causing the current to vary in the same manner. To obtain definitive input data relative to the electrothermal gasifier bed, the voltage and current were monitored during tests. Oscillograph traces were obtained at various time intervals to record the instantaneous changes in the voltage and current. A typical oscillograph trace of the voltage and current appears in Figure 4a.1-17.



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Figure 4a.1-16. RUN EG-48 RESULTS SCALED TO MAXIMUM REQUIREMENTS



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Figure 4a.1-17. OSCILLOGRAPH TRACES OF VOLTAGE AND CURRENT DURING RUN NO. 53

The non-sinusoidal character of the current is readily apparent; the extreme in the ratio of maximum to minimum values of current is 4. In contrast to the arc furnace operation, the current trace is continuous and arcs are not present on a continuous basis. This absence of continuous short-circuit operation makes the load more attractive from the viewpoint of less interference with the main supply source, and with other loads connected to the main load source.

Data from tests in the 6-inch electrothermal gasifier bed were used to develop techniques for obtaining an empirical relationship from voltage-current characteristics, and to obtain scaling factors applicable to larger systems of the same configuration. In the concentric electrode configuration (Figure 4a.1-18) the resistivity of an electrothermal gasifier varies according to -

$$\rho = kJ^n \quad (4)$$

neglecting end effects, the electric field intensity in the bed is -

$$E = \rho J = kJ^{n+1} \quad (5)$$

and the voltage between electrodes is-

$$V = + \int_{r_i}^{r_o} kJ^{(n+1)} dr \quad (6)$$

since

$$JA = J_i A_i \quad (7)$$

or

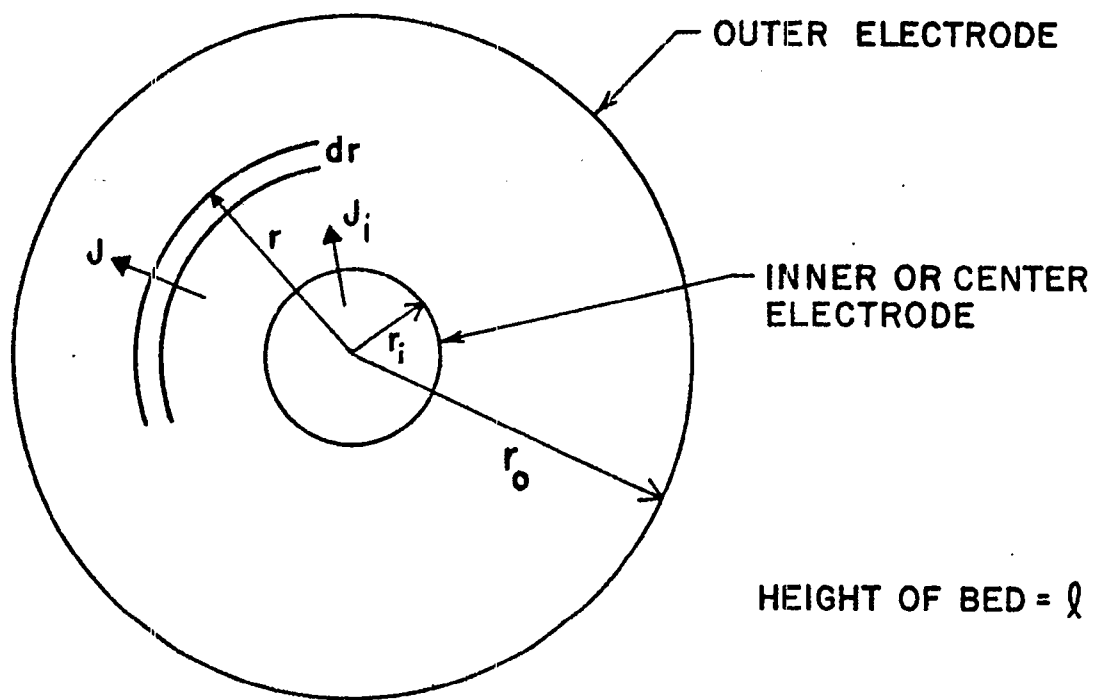
$$J = J_i \frac{A_i}{A} = J_i \frac{r_i}{r} \quad (8)$$

Then

$$V = + \int_{r_i}^{r_o} k (J_i)^{n+1} \left(\frac{r_i}{r}\right)^{n+1} dr \quad (9)$$

in which case

$$V = + \frac{kJ_i^{n+1} r_i}{n} \left[1 - \left(\frac{r_i}{r_o}\right)^n \right] \quad (10)$$



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Figure 4a.1-18. CROSS-SECTION OF A TYPICAL CONCENTRIC ELECTRODE CONFIGURATION

substituting $I_i = J_i A_i$,

$$V = + \frac{kr_i}{n} I_i^{n+1} \left(\frac{l}{2\pi \ell r_i} \right)^{n+1} \left[1 - \left(\frac{r_i}{r_o} \right)^n \right] \quad (11)$$

Resistance is defined as $R = V/I$, so that

$$R_{BED} = \frac{V}{I_i} = \frac{kr_i}{n} I_i^n \left(\frac{l}{2\pi \ell r_i} \right)^{n+1} \left[1 - \left(\frac{r_i}{r_o} \right)^n \right] \quad (12)$$

which, for a given bed, reduces to

$$R_{BED} = C I_i^n \quad (13)$$

where C is a constant defined as -

$$C = \frac{kr_i}{n} \left(\frac{l}{2\pi \ell r_i} \right)^{n+1} \left[1 - \left(\frac{r_i}{r_o} \right)^n \right] \quad (14)$$

For a given bed, the voltage, Equation 11, reduces to a simple expression -

$$V = k' I^{(n+1)} \quad (15)$$

where k' , a constant, is -

$$k' = \frac{kr_i}{n} \left(\frac{l}{2\pi \ell r_i} \right)^{n+1} \left[1 - \left(\frac{r_i}{r_o} \right)^n \right] \quad (16)$$

The bed resistance, Equation 12, also can be given in terms of current density. The correct expression is -

$$R_{BED} = \frac{k}{2\pi n \ell} J_i^n \left[1 - \left(\frac{r_i}{r_o} \right)^n \right] \quad (17)$$

where J_i is the current density at the inner electrode. For $n \cong -1/2$, Equation 15 becomes -

$$R_{BED} = \frac{k}{\pi \ell} \frac{1}{J_i^{1/2}} \left[\left(\frac{r_o}{r_i} \right)^{1/2} - 1 \right] \quad (18)$$

The constants k' and n were determined from a voltage-current (V-I) characteristic for a given bed. Data from the 6-inch electrogasifier bed are presented in Figure 4a.1-19. The data were taken at steady-state operating conditions when the voltage would be varied over a wide range and both voltage and current traces acquired on the oscillograph at the different settings. The time required to obtain the data was only several minutes and the effect on the steady-state operation of the system was negligible. An averaged straight line through the available V-I characteristic data yields a k' of 15.7 and an n of -0.468 where $r_i = 0.75$ inch, $r_o = 3$ inches, and $\ell = 24$ inches. By using these values, k is found to be 129 (ρ in ohm-in. and J in A/sq in.), so that from equation 4

$$\rho = 129 \frac{1}{J^{0.468}} \text{ ohm-in.} \quad (19)$$

These results were obtained from three tests of the 6-inch electrogasifier bed. Based on all tests of the 6-inch electrogasifier bed, the constant k was determined to be on the average 87 (ρ in ohm-in. and J in A/sq in.).

4a.1.9.2 Resistance

The overall resistance of the system was obtained by metering the voltage output of the d-c generator, the average current flow in the circuit and applying Ohms Law ($R = \frac{V}{I}$). The average overall resistance during the steady state operating period varied from 0.5 to 2.0 ohms but was normally between 0.5 and 1.0 ohms. Tests with the various char materials all reached steady-state operation as illustrated in Figure 4a.1-20.

4a.1.9.3 Resistivity

The resistivity of the char bed during the steady-state period of the tests was then calculated by inserting the measured overall resistance of the system and electrode geometry into equation 3.

The relationship of resistivity to the current density of the center electrode closely followed those of previous HYGAS-related studies at high pressure but at lower current densities (Figure 4a.1-4). Figure 4a.1-21 illustrates the trend observed in tests at 1000 pounds per square inch (gage) and temperatures from 1750° to 1900° F.

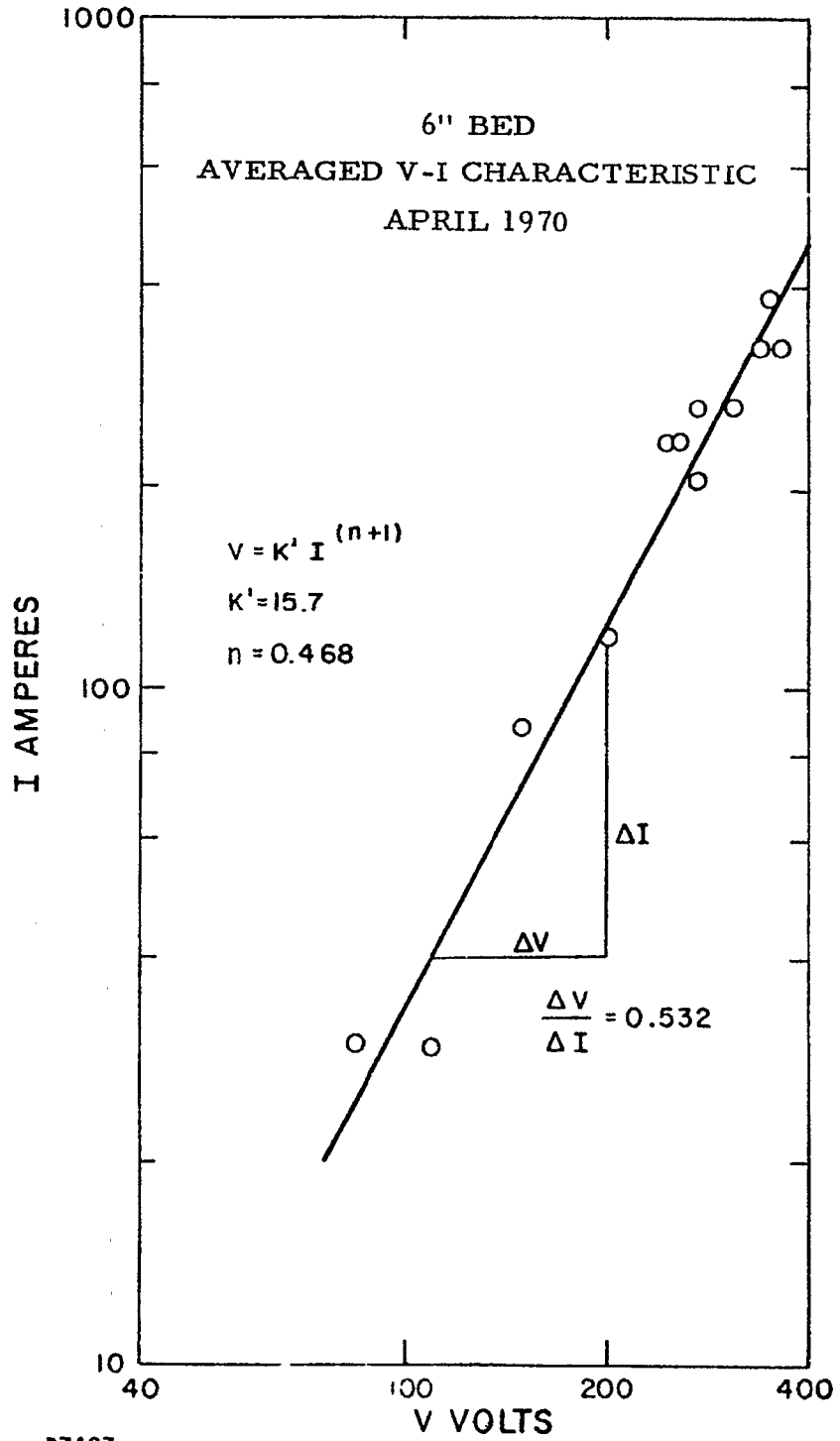
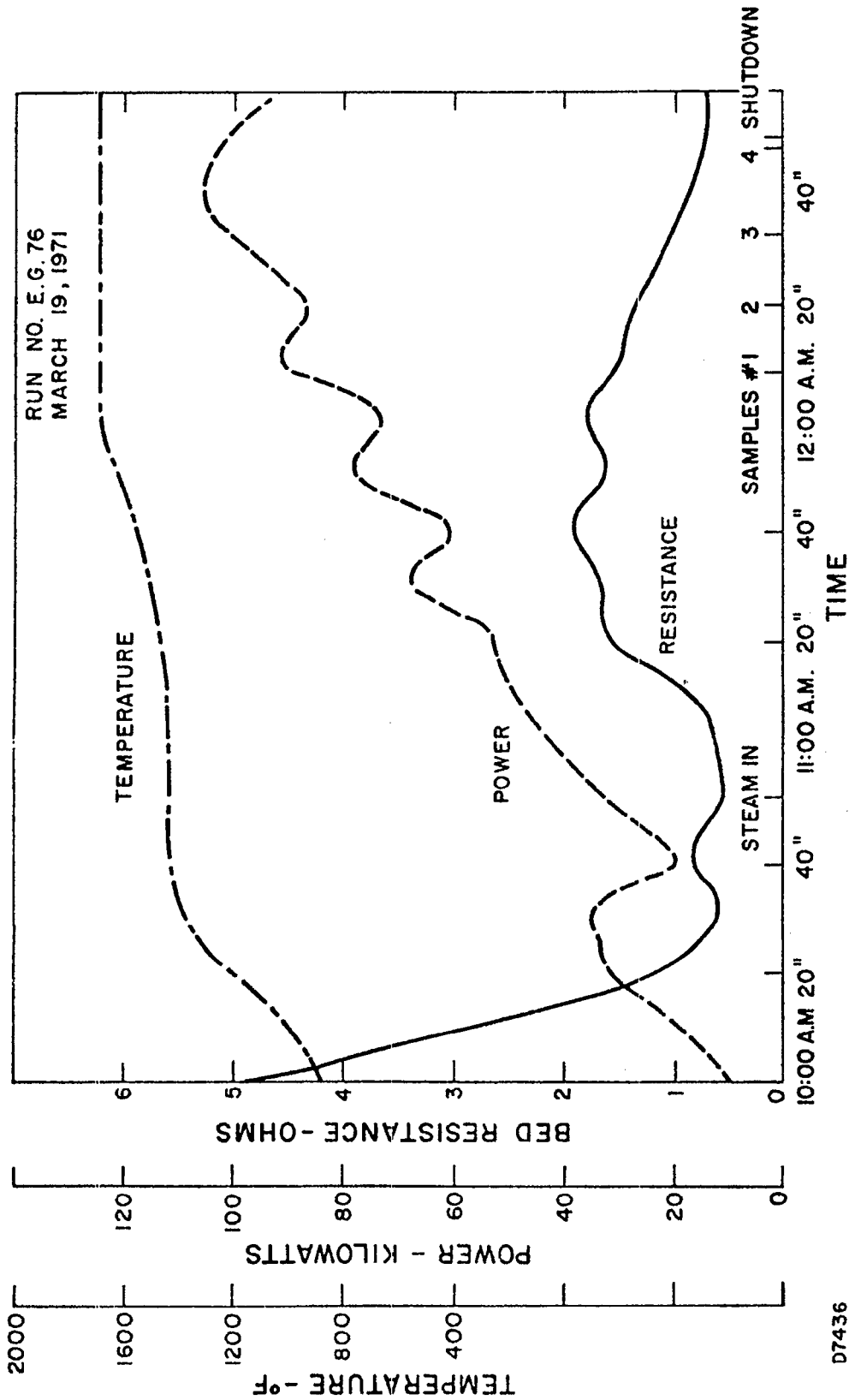
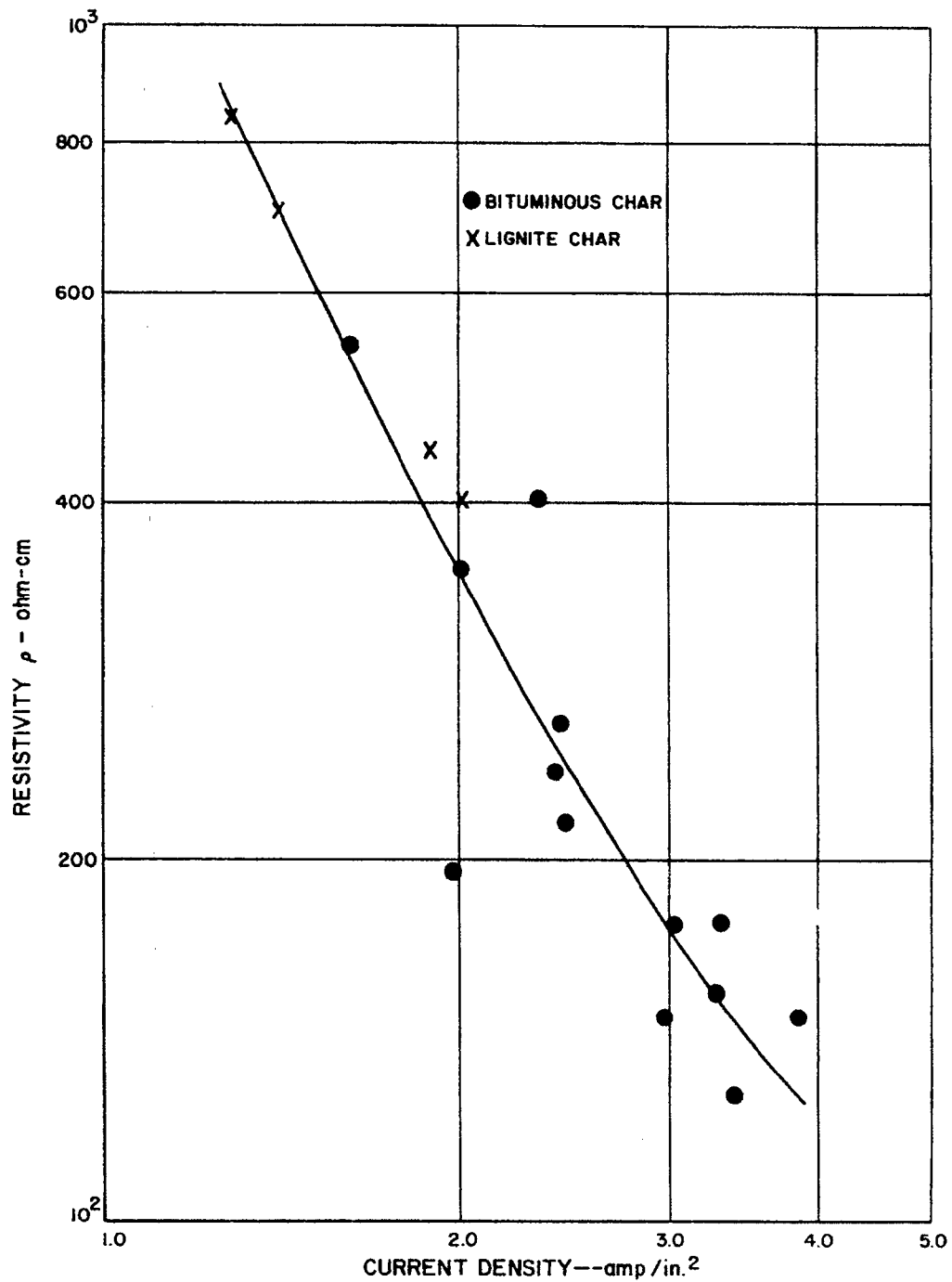


Figure 4a.1-19. VOLTAGE CURRENT CHARACTERISTIC OF THE 6-INCH ELECTROTHERMAL GASIFIER BED



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Figure 4a.1-20. VARIATION OF BED RESISTANCE, INPUT POWER AND BED TEMPERATURE DURING TEST NO. 76



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Figure 4a.1-21. EFFECT OF CURRENT DENSITY ON RESISTIVITY AT 1000 lb/sq. in. PRESSURE AND TEMPERATURES OF 1700° to 1915°F