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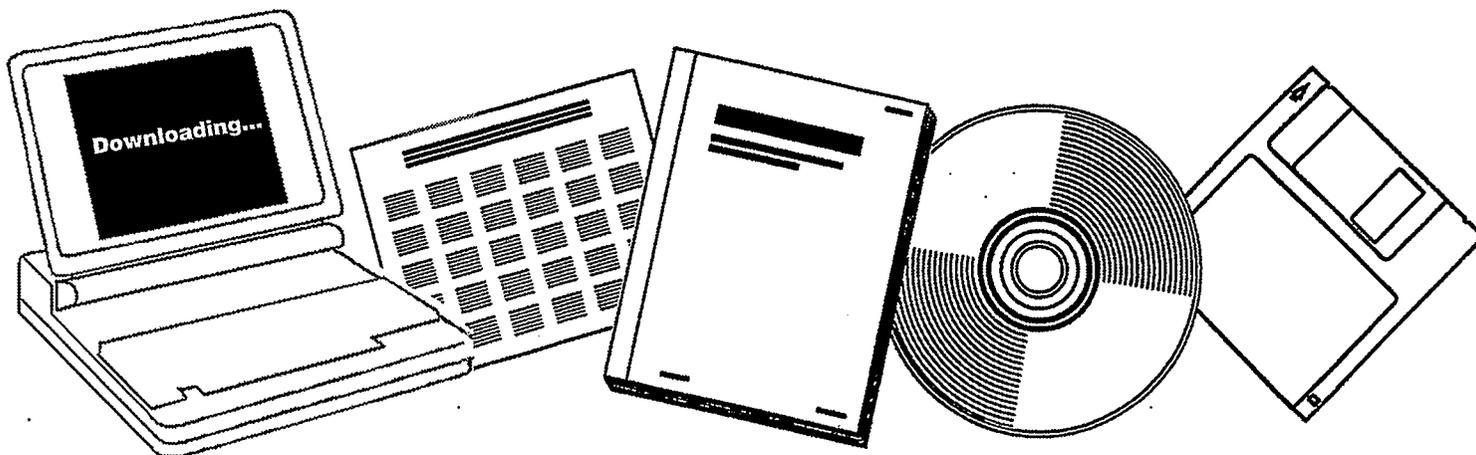
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**HYGAS: 1964 TO 1972. PIPELINE GAS FROM  
COAL--HYDROGENATION (IGT HYDROGASIFICATION  
PROCESS). VOLUME 1**

INSTITUTE OF GAS TECHNOLOGY, CHICAGO,  
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JUL 1975



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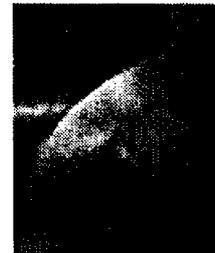
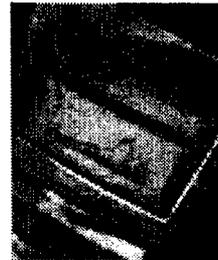
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HYGAS<sup>TM</sup> : 1964 TO 1972  
PIPELINE GAS FROM COAL — HYDROGENATION  
( IGT HYDROGASIFICATION PROCESS )

Final Report

by

**IGT Process Research Division**

Prepared for

**ENERGY RESEARCH AND DEVELOPMENT ADMINISTRATION**

**Washington, D.C. 20545**

July 1975

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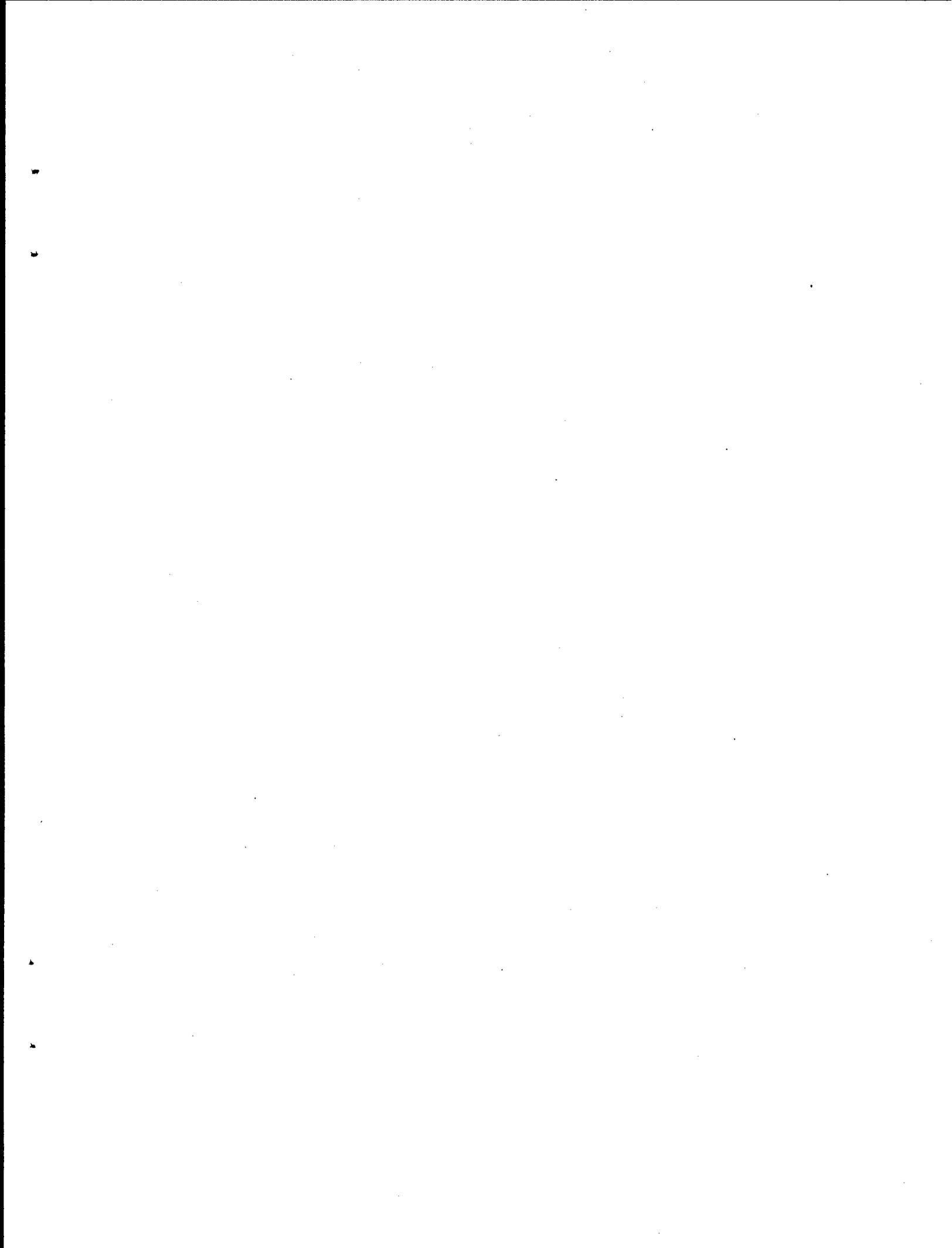


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## 1.0 Introduction to the Report

OCR R&D Report No. 22 is a comprehensive presentation of activities conducted by the Institute of Gas Technology (IGT) for the Department of the Interior, Office of Coal Research (OCR) and its associated sponsor, the American Gas Association (A. G. A.), concerning **HYGAS™ process research** under the following contracts: OCR Contract No. 14-01-0001-381, dated July, 1964; 14-01-0001-381(1), dated June, 1967; and 14-01-0001-381(2), dated March, 1972. The work was performed in the period of July 1964 to September 1972. Earlier work from which the HYGAS\* process concept emerged was privately sponsored, principally by the A. G. A., and is reported in IGT Research Bulletin 39. The work reported in this OCR R&D Report No. 22 is continuing under the U. S. Energy Research & Development Administration (ERDA) Contract No. E(49-18)-1221. This report was produced by the Process Research Division of IGT in compliance with contract directives.

A comprehensive yet flexible outline form was adopted early in the preparation of this report to permit simultaneous independent work on each of the various parts, with independent publication if desired. The principal parts of the report are:

### Volume 1

Tables of Content, Lists of Figures, and Lists of Tables

Part I: Summary (including Bibliography)

Part II: Hydrogasification in a 4-Inch PDU

### Volume 2

Part III: Pilot Plant Development

Part IV: Hydrogen Generation

### Volume 3

Part V: Methanation

Part VI: Coal and Char Characterization

Part VII: Coal Pretreatment in a 10-Inch PDU

### Volume 4

Part VIII: Commercial Plant Design

Part IX: Process Economics

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\* HYGAS is the IGT acronym for HYdroGASification, and is a registered trademark.

Because the outlining was established before the number of volumes could be determined, the Roman numeral that identifies each part in the above list — rather than the volume number — provides the base-reference key for all divisions of the report text. Thus, section 3.2 is the second major section of Part III; a decimal system is utilized with increasing numbers of decimal digits and increasing indentation to label subordinate divisions of text. With the decimal index, a portion of the report referenced only by a section number can be quickly located simply by scanning the left margin of text, if desired, without turning to the table of contents. The content in most parts of the report is limited to a single major area of work; however, Parts IV and VI each pertain to several different (although related) work areas. In parts IV and VI, therefore, the subsections are delineated by a letter code; for example, Part VI is divided into subordinate Parts 6a, 6b, 6c, and 6d.

The report part number also provides the base-reference for all pagination, illustrations, tables, and appendixes. Thus, page 1-23 is the twenty-third page of Part I, Figure 8-22 is the twenty-second figure of Part VIII, Table 5-46 is the forty-sixth table in Part V, and Appendix 6c-C is the third appendix of subordinate Part 6c.

Each volume starts with a brief list of the parts included therein. The complete table of contents and complete lists of figures and tables for the entire report appear at the front of Volume 1. In addition, the appropriate complete table of contents, list of figures and list of tables are repeated at the front of each part of the report.

While most nomenclature is rather straightforward, the designations of experimental run data (aside from the run numbers) are often preceded by codes. These letter codes, derived from various IGT research areas, are as follows:

HT	-	High Temperature
EG	-	Electrothermal Gasifier
EG-O	-	Electrothermal Gasifier/Oxygen
SO	-	Steam Oxygen
OH	-	Oxygen Hydrogasifier
FP	-	Fluidized-bed Pretreatment
F-OH	-	Feed (char), Oxygen Hydrogasifier
R-OH	-	Residue (char), Oxygen Hydrogasifier
R-HT	-	Residue (char) — High Temperature
R-EG	-	Residue (char) — Electrothermal Gasifier
LS	-	Life Study (catalysis)
LT	-	Life Test (catalysis)
HS	-	Hydrogen Sulfide test (catalysis)
TL	-	Test Laboratory (heat of reaction)

### 1.1 Introduction to the Summary

This summary is comprised of the individual summaries that precede each part of this report.

## 1.2 Summary of Hydrogasification in the 4-Inch PDU

During the contract period reported in this document, starting in 1964 and ending in 1972, more than 250 coal hydrogasification tests were performed under a variety of conditions and utilizing many types of coal in combination with a variety of gaseous feeds.

The work was performed in a 4-inch-diameter reactor termed the hydrogasification process development unit (PDU), which is described in its various configurations herein. The results of this PDU hydrogasification work helped to confirm early HYGAS concepts and played a significant role in shaping criteria for design of the HYGAS pilot plant reactors.

In the early phases of operation and shakedown of the demonstration unit, a low-temperature bituminous coal char of uniform composition was used. During the investigative phase of the hydrogasification program, 14 different coals were evaluated for the effect of pretreatment operations and/or char composition on the hydrogasification results. The coals were selected to give a range in rank from a lignite to a low-volatile bituminous as well as for three different high-volatile bituminous coals to determine variation in performance of coals due to origin. FMC Corporation Project COED char was also investigated for comparison.

Early moving-bed experiments were conducted with -35+80 USS mesh char for low throughputs and -10+40 USS mesh for high throughputs of coal and gas feeds. The coarser feed allowed higher gas velocities without fluidizing the char bed. For fluid bed tests, the particle size was extended to cover the whole range of char feed. The -10+80 mesh size was used for all fluidized-bed tests.

Initially, the hydrogasification program utilized a moving-bed configuration which offers a number of advantages. At the top, highly reactive fresh coal is in contact with the product gas which results in high methane equilibrium concentrations in the product gas. Less reactive partially gasified char is in contact with hydrogen-rich feed gas at the bottom of the bed which promotes the rates of both the carbon-hydrogen and carbon-steam reactions.

Typically, hydrogasification of lightly pretreated Ireland Mine bituminous coal with hydrogen and steam in a moving bed resulted in up to 38.5% carbon gasification and a product gas of 495 Btu per standard cubic foot. Results with Montour No. 4 mine pretreated coal and a North Dakota lignite are 28% and 57% carbon gasified and 576 and 511 Btu per standard cubic foot of product gas, respectively.

It was found, using volatile matter content as an index of pretreatment severity, that pretreated coal with between 24 and 26% volatile matter can be processed without agglomeration in a 4-inch diameter reactor. It may be possible to feed coals with less pretreatment - or even raw coal - in a larger diameter reactor.

Experience was gained from these tests in adjustment of feed tube size, length and location; the amount of nitrogen purge-gas required to pass through the tube; and the start-up sequence necessary for a successful operation.

Two-stage hydrogasification was simulated in sequence, by feeding the pretreated coal in the low-temperature stage and partially gasified coal from the low-temperature stage to the high-temperature stage. This is realistic except that the partially gasified coal was fed to the second (high-temperature) stage at ambient temperature instead of at temperatures between 1200° and 1400° F. All the tests were conducted at minimum hydrogen/coal ratio to yield about 50% carbon gasification, which provides sufficient residual carbon for hydrogen generation in a subsequent operation.

Because of the low temperature in the first stage, steam-carbon reaction is not expected in the first stage. The carbon-oxides formed in this stage come from the organic oxygen in the coal. However, steam does play an active role in low-temperature gasification. Experimental results indicate that steam seems to suppress the release of oxygen from coal as water and forces it to leave as carbon oxides. This is desirable because it reduces hydrogen loss and carbon oxides can be further converted to methane by catalytic methanation.

It has been well established that the hydrogenation of the volatile matter in coal proceeds very rapidly and yields methane concentrations higher than the equilibrium value in a  $\beta$ -graphite-hydrogen system. The first-stage hydrogasification demonstrated this type of reaction as shown by the observed rapid rate of reaction and lack of equilibrium hindrance to methane formation in this stage. The partially gasified coal (in the high-temperature stage) was less reactive at temperatures from 1700° to 1900° F.

On comparing the integral methane formation rates with those reported by others, it was found that 1) the rate of methane formation for pretreated Pittsburgh coal is not slowed by the presence of methane in the feed gas; 2) the pretreated coal is quite reactive, probably due to its high volatile matter content as compared to others; and 3) in the high-temperature stage, partially gasified coal gave rate constants quite similar to those obtained with Disco char and residual Australian brown coal, both of which had very little volatile matter.

The carbon-steam reaction was significant only at temperatures above 1700° F, and reaction rate was found to increase with temperature. At the hydrogasification temperature utilized, this reaction is expected to be substantially removed from equilibrium. The carbon-steam reaction is important not only as a source of generating in situ hydrogen, but also as a built-in temperature controller since it absorbs the heat generated by exothermic carbon-hydrogen reaction.

Gas samples collected at different levels indicated that in the high-temperature test the reaction was practically complete in the lower half of the bed, whereas in the low-temperature test the bulk of the reaction took place in the free-fall zone and at the top of the bed. This is to be expected because the high-temperature reaction is equilibrium-limited and apparently attains its limit in a relatively short contact time, and the low-

temperature reaction is extremely rapid, requiring only a matter of seconds for completion (the residual carbon being less reactive at the low temperature). In view of this, a likely hydrogasifier configuration would incorporate a low-temperature zone for the rapid reactions followed by a high-temperature zone for the slower reactions.

Free-fall tests were conducted to study coal conversions at short residence times. The coal residence times varied from 8 seconds for 60 mesh to 41 seconds for 200 mesh particles. The coal feeds included medium and low-volatile-content bituminous coals, a subbituminous coal and two different lignites. The tests indicated:

1. Most of the steam reacts with char rather than gaseous hydrocarbons, and hence the steam decomposition is dependent on the char residence time.
2. It is possible to hydrogasify a highly caking coal (Pocahontas No. 4 Seam) in free-fall without agglomeration.
3. It is feasible to convert lignite (up to 37%) at high pressures by pyrolysis in a free-fall.

A major operational restriction to moving-bed operation was the limit it set on coal and gas feed rates. Fluidized-bed operation would overcome this and increase the hydrogasification capacity significantly. Fluidized-bed operation, with its solids backmixing, would also eliminate the reaction gradient and yield uniform temperature and composition for the char bed.

Fluidized-bed tests were conducted at selected conditions on eight bituminous coals, three subbituminous coals and two lignites. Two-stage hydrogasification was also simulated in two sequential tests, as in the previous moving-bed tests. The feed gas was varied from hydrogen-steam mixtures to synthesis gas plus steam, mixtures of hydrogen, methane and steam, and mixtures of synthesis gas, methane and steam to simulate the two-stage hydrogasifier.

The following conclusions can be drawn from the fluidized-bed tests in the 4-inch-diameter reactor:

1. As shown in the free-fall tests, high-, medium-, and low-volatile bituminous coals can be successfully hydrogasified in a lean-phase mode before direct injection into a fluidized bed. This indicates that pretreatment of these coals may be eliminated by a two-step lean-phase process followed by dense-phase fluidization. Lignite and subbituminous coals generally need no pretreatment; one subbituminous coal (Colorado Laramie No. 3 seam subbituminous) performed marginally without a pretreatment. There is no significant difference in hydrogasification results for medium and high-volatile-content bituminous coals.
2. Subbituminous coals are more reactive than pretreated bituminous coals but somewhat less active than the lignites. For both the subbituminous coal and the lignite feeds, hydrocarbon yields were similar. The increased carbon gasification with lignite was due mainly to the increased yield of carbon oxides and oil products.

3. Hydrogasification of FMC Corporation's Project COED char showed that this char has a lower reactivity than pretreated or partially-gasified bituminous coal from Pittsburgh No. 8 seam, Ireland Mine.

The use of synthesis gas instead of hydrogen in the feed gas to the hydrogasifier is of interest as it would eliminate the need for carbon monoxide shifting and carbon dioxide removal following the hydrogen production step. Results of the tests using synthesis gas-steam mixtures show that for bituminous coals the reaction rates are quite sensitive to the hydrogen partial-pressure in the system. The use of synthesis gas in place of hydrogen requires operating adjustments in order to maintain the necessary conversion levels of carbon and steam. These adjustments may consist of 1) longer residence time, 2) higher steam concentration to increase water-gas shift reaction, 3) higher synthesis gas-to-coal ratio, 4) higher system pressure to increase hydrogen partial pressure, and/or 5) multistage contacting to improve countercurrency, or combinations thereof. However, with lignite, the full benefit of hydrogasifying with a synthesis gas instead of hydrogen can be realized without any offsetting penalty. A beneficial effect of using synthesis gas for lignite hydrogasification is the apparent suppression of oil production.

The reaction of subbituminous coal to hydrogasification with synthesis gas is similar to that exhibited by bituminous coals. In another test, FMC char also yielded lower carbon gasification with synthesis gas than that with hydrogen.

The principal advantages of operation at a lower pressure are reduced capital investment and easier solids handling. These have to outweigh the reduced methane production and carbon conversion to be expected at lower pressure. Tests with high-volatile bituminous coal at 500 psig indicate that the product gas does indeed have lower methane concentration at lower pressure (27.5% versus 36.9%) for similar carbon conversions. For lignite the hydrocarbon yield at 500 psig was only 86% of that at 1000 psig, while the carbon oxides yield was 115% of that at higher pressure. Sixty-four percent less carbon in lignite was converted to oil at 500 psig. The use of synthesis gas instead of hydrogen did not unduly affect the carbon conversion at lower pressure for lignite.

For subbituminous coal, the carbon gasification, hydrocarbon yield and product gas heating values were significantly greater at 1000 psig than at 500 psig. The carbon gasified at 500 psig compares favorably with that at 1000 psig when synthesis gas and steam are used as feed gas.

#### 1.2.1 Product Gas Composition and Trace Components

The major constituents of the product gases produced by the hydrogasification of coal and lignite with hydrogen and steam in the PDU tests discussed are: methane, carbon oxides (carbon monoxide and carbon dioxide), and hydrogen. Methane yield is determined by the degree of coal conversion and may also be controlled by the reactor temperature that sets the equilibrium composition of the product gas. The amount of carbon oxides formed is dependent on the oxygen content of the coal feeds and the steam-to-coal ratio. Feeds high in oxygen content such as subbituminous coal and lignite, when gasified, produce more carbon oxides than bituminous coals produce at

similar conditions. In addition, the higher the steam-to-coal ratio, the greater will be the carbon oxides production, other conditions being equal. The ratio of carbon monoxide to carbon dioxide is set by the water-gas shift reaction, which is temperature controlled. The hydrogen content of the product gas is determined largely by the quantity of unreacted hydrogen remaining after the reaction. The concentration of hydrogen is related directly, but not linearly, to the feed hydrogen-to-coal ratio. As the stoichiometric hydrogen-to-coal ratio is increased, hydrogen concentration in the product gas tends to increase at a greater rate.

In addition to methane, small quantities of other saturated hydrocarbon are produced. Typical concentrations of these components are, ethane, 1.5%; propane, 0.5%; and butane, 0.2%. The principal aromatic in the product gas, generally less than 1%, is benzene.

A portion of the nitrogen in the coal is gasified and a substantial part, if not all, of it appears in the reactor product gas as ammonia, which hydrolyzes in the water condensate.

While there is considerable scatter in the data, it is evident that sulfur gasification tends to increase as gasification proceeds and that the sulfur is preferentially gasified, i. e. the percent conversion of feed sulfur is always higher than percent conversion of coal on a moisture- and ash-free basis. Most of the sulfur appears in the reactor product gas as hydrogen sulfide, accompanied by small amounts (probably less than 200 ppm) of carbonyl sulfide (COS) and traces of organic sulfur compounds.

### 1.2.2 Char Compositions

Char rates and compositions reflected the degree of conversion of the feeds to gaseous and liquid products. Volatile matter contents of bituminous coals were generally reduced to 3% or less, and to less than 7% for subbituminous coal and for lignite. The reduction in hydrogen content tended to parallel the reduction in volatile matter content. Hydrogen concentration in the residue was reduced to less than 1.5% for all feeds. Gasification of the bituminous coals resulted in nearly complete consumption of the oxygen in the coal to produce carbon oxides and water. The residues produced from subbituminous coal and lignite gasification still contained from 3 to 6% oxygen by weight. In general, sulfur concentrations were reduced up to 50% in bituminous coal chars, and more than 50% in subbituminous and lignite residues. Carbon concentration, in all cases, was significantly higher in the residue than in the feed.

There was no observable trend in the change in size distribution of the coal feeds after hydrogasification. Any breakdown in particle size due to attrition, or fracture, induced by the reaction, appeared to be balanced by particle growth due to coalescence.

Gasification of the coals and lignite resulted in a sizeable reduction in the bulk density. Lightly pretreated bituminous coal bulk densities were reduced from a range of 20 to 25 pounds per cubic foot, to a range of 15 to 20 pounds per cubic foot. Bulk densities of dried, untreated subbituminous coal and lignite were of the order of 45 pounds per cubic foot. After gasification, the subbituminous coal bulk density was reduced to 28 pounds per cubic foot, while the lignite bulk density was reduced to 20 pounds per cubic foot.

### 1.2.3 Liquid Products

Condensed liquids resulting from the hydrogasification of coals and lignite consisted mainly of water and a light oil fraction. Small quantities of ammonia and unidentified sulfur compounds were generally dissolved in the liquids. The relative quantities of water and oil were dependent mainly on the amount of steam fed and steam decomposed, and on the nature of the coal feed. Feeds high in volatile matter content, such as subbituminous coal and lignite, produced more oils than bituminous coals, other conditions being equal. Oil production was also influenced by reactor temperature, being higher at lower temperatures than at higher temperatures.

Specific gravity of the by-product oils was in the range of 1.001 to 1.010. Typically, the oil composition was 90 weight percent carbon and 6 weight percent hydrogen, the remainder being small quantities of oxygen, nitrogen, and sulfur. The principal compounds in the oil are benzene and toluene.

### 1.3 Summary of Pilot Plant Development

The preliminary engineering design of the HYGAS pilot plant was performed by Bechtel Corporation in association with IGT.

The final design and construction of the HYGAS pilot plant was performed by Procon, Inc., a division of UOP, in association with IGT.

Detailed discussion in Part III covers process chemistry, preliminary plant design, design modifications and construction, and start-up operations. Problems are delineated as well as their solutions, and findings and recommendations applicable to other coal hydrogasification plant designers are informally presented.

The first seven start-up test runs are reported in detail; however, the limited amount of operational data obtained during the contract period made it inadvisable to present that material here. Instead, with the consent of OCR, operational data from the period reported here will be presented in more meaningful perspective in reports of the subsequent HYGAS contract sponsored by OCR with the support of the A. G. A. (OCR Contract No. 14-32-0001-1221, now ERDA Contract No. E(49-18)-1221.)

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°-1850° 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 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, 65-70% 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%. If the process were operated at a lower pressure, more methane would need to 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,



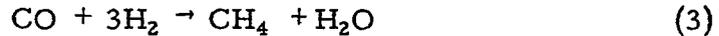
is strongly exothermic, while the coal-steam reaction,



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.

In the pilot plant, the hydrogasifier reactor vessel is 135 feet high; the outer pressure shell has a 5.5-foot inside diameter. The slurry is sprayed into the dryer, a 2.5-foot-diameter, 10-foot-high fluidized drying bed. The sensible heat in the gaseous reaction products from the later stages vaporizes 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 — the low-temperature reactor. At the top of the lift line, the gas and coal disengage. The gas moves up to vaporize the oil in the slurry-drying bed. 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, IGT believes, raw caking coal can be fed directly to the reactor without pretreatment. Eliminating pretreatment can reduce gas cost significantly.

The remainder (or all) of the partially reacted coal flows by gravity to the second-stage hydrogasifier — the high-temperature reactor. The second-stage bed is 2.5 feet in diameter, is lined with refractory, and is 15 feet deep. 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\* below. (Or, alternatively, the hydrogen-rich gas may flow from the steam-iron reactor or the electrothermal generator.)

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 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.

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\* An electrothermal gasifier was built in the period reported; a steam-oxygen gasifier was installed after this contract period; a steam-iron gasifier is under construction in mid-1975.

From this reaction stage, the char goes to the hydrogen-producer gasifier, 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 high-temperature stage gasifier. 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 is recycled.

The composition of gas synthesized in the two principal reactor stages, and passed upward through the slurry dryer at the top of the hydrogasification reactor, depends on the type of hydrogen producer, as shown in the following table. In addition to these major components, the gas contains the slurry oil, coal dust, and trace constituents such as ammonia and hydrogen cyanide.

#### GAS COMPOSITION LEAVING HYGAS REACTOR

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

The gas mixture delineated is at 600°F; it 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.

\* Installed in the base of the existing HYGAS reactor vessel after the close of this contract period.

Calculations indicate that the purified gas entering the methanation section of a commercial plant would typically have the composition tabulated below. 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.

#### GAS COMPOSITION ENTERING THE HYGAS METHANATOR

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

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 to slightly above 3\*. High pressure and low temperature favor completion of the methanation reaction *i. e.* — in the methanator. 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.

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\* A water-gas shift section would be installed for this purpose in a commercial operation.

#### 1.4 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. By using a mixture of hydrogen and steam instead of hydrogen alone, the heat released by one principal reaction in the reactor is absorbed in situ by a second principal reaction, resulting in:

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

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 electrothermal 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.

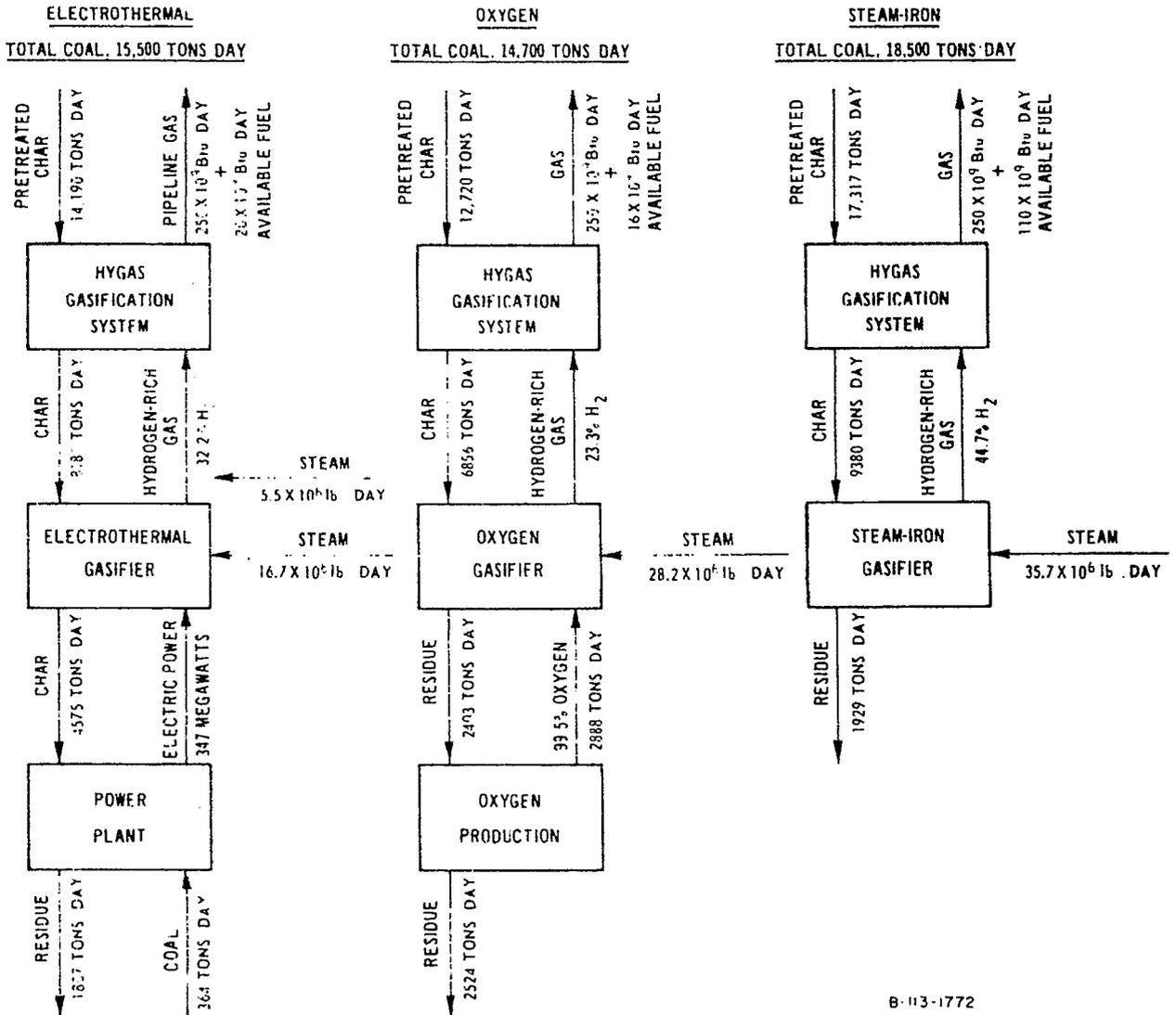
##### 1.4.1 Hydrogen for the HYGAS Process

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 1-1.

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

\*\* Following the contract period reported here.



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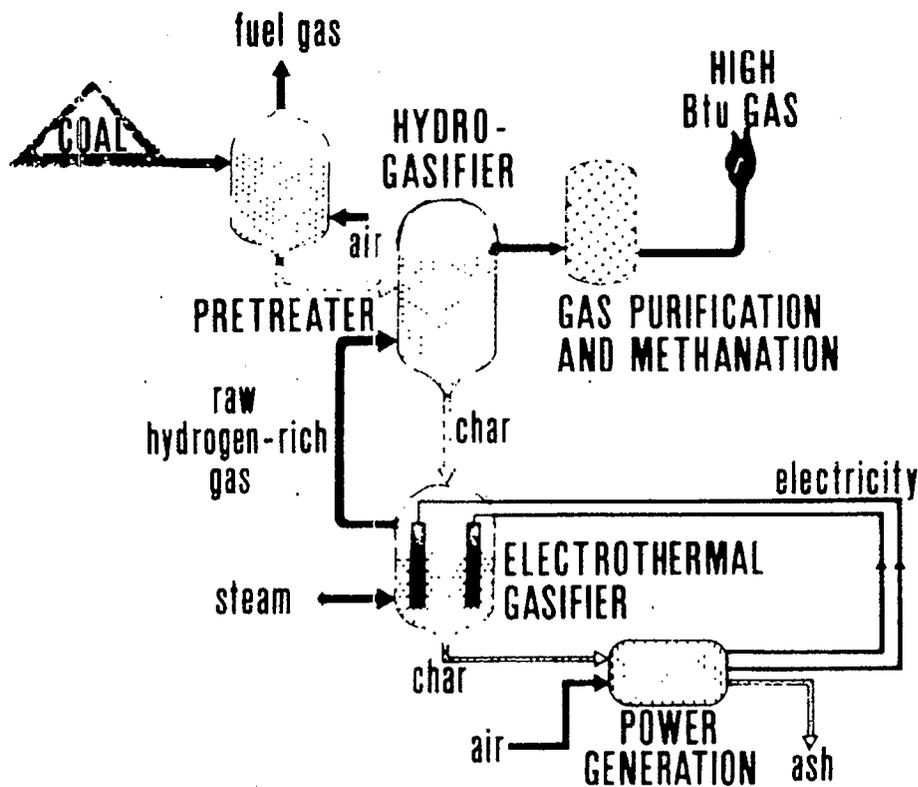
Figure 1-1. EFFECT ON THE IGT HYGAS PROCESS FROM USING THREE DIFFERENT SYSTEMS FOR PRODUCING HYDROGEN-RICH GAS: PLANT CAPACITY IS  $250 \times 10^9$  Btu/DAY OF PIPELINE-QUALITY GAS

#### 1.4.2 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 investigate initially 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 1-2.)

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 acid<sup>1</sup>, processes for the calcination of petroleum coke<sup>2</sup> and various metallurgical applications<sup>3</sup> have emerged and prospects for future developments are numerous.<sup>3, 4</sup>

Preliminary studies<sup>5, 6</sup> 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



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Figure 1-2. THE HYGAS PROCESS SHOWN WITH AN ELECTROTHERMAL GASIFIER TO PRODUCE HYDROGEN-RICH GAS

showed that the electrical characteristics of a spent char system are similar to those of the other materials<sup>7, 8, 9</sup> 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.

#### 1.4.3 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, to be published.

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.

#### 1.4.4 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 CO<sub>2</sub> and methane formation supply the heat for the steam-char reaction that produces H<sub>2</sub> 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.

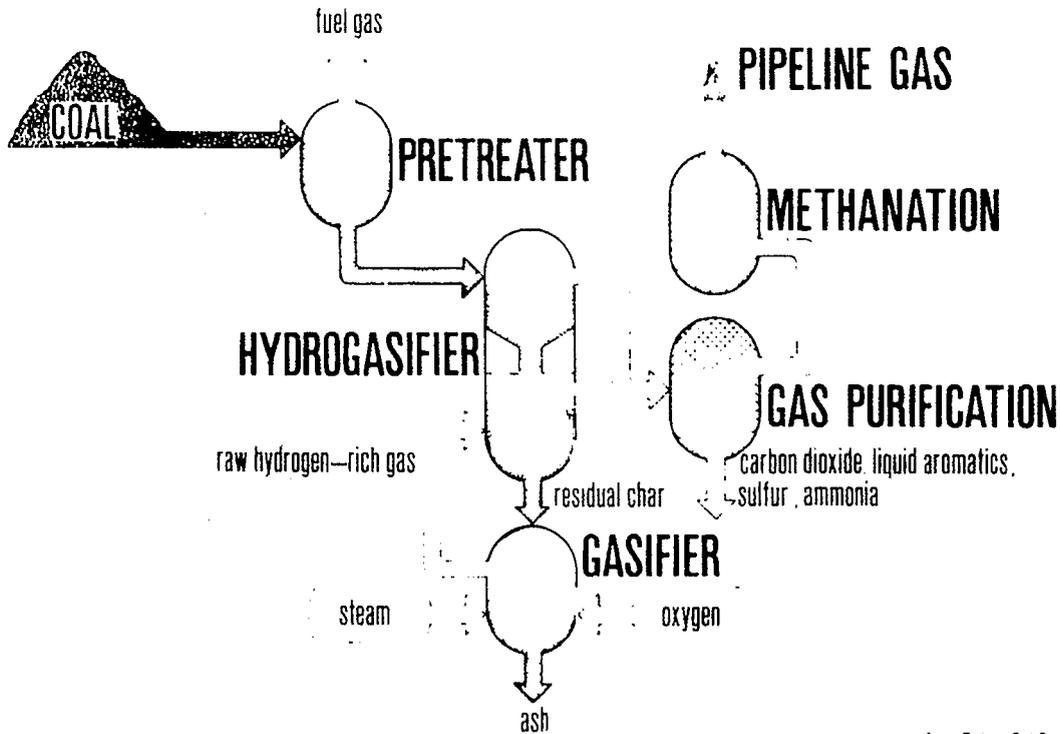
The basic questions to be resolved in the steam-oxygen program are —

- 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 1-3) 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 1-3. 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.

#### 1.4.5 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.

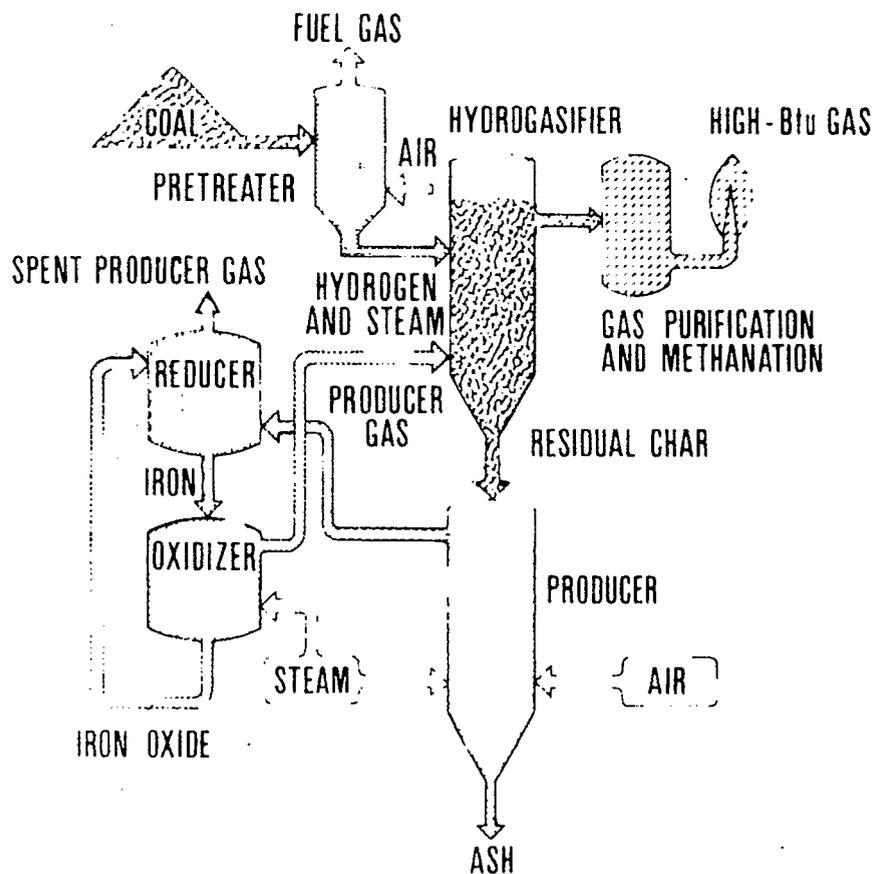
The steam-iron process for making hydrogen is very old. Historically, the early process was operated at atmospheric pressure using two beds of iron solids. One bed was reduced from iron oxide to iron by a suitable gas while the second bed was being oxidized by steam from iron to iron oxide.

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 1-4 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/CO<sub>2</sub> and H<sub>2</sub>/H<sub>2</sub>O 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 1-4. 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.

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\* 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.

1.4.6 References Cited

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## 1.5 Summary of Methanation

In this section, the work carried out on the evaluation of catalysts for the methanation of coal gasification effluent gas streams (to raise the heating value to the level of pipeline gas) is reported. Both the conventional packed-bed reactor (PBR) and a continuous-stirred-tank reactor (CSTR) used in the study, along with the associated instrumentation, are described. The "perfect-mixing" zone for the CSTR is established experimentally. Operation in the "perfect-mixing" zone in the CSTR eliminates the temperature and concentration gradients, and permits an easier evaluation of the kinetics of a chemical reaction.

A nine-step test program was developed to evaluate various catalysts on the same basis. Since 1964, a total of 37 commercially available and newly developed catalysts have been tested. Not all the catalysts were subjected to the complete test program; evaluation was stopped at the step where a catalyst failed. Performance data for each catalyst tested are available, if not reported herein.

The catalyst properties evaluated were activity, physical strength, upper and lower temperature limits, resistance to poison, selectivity for methanation reaction and the life performance. No single catalyst satisfies all the requirements of an ideal catalyst. For example, some activity may have to be sacrificed for additional physical strength, or the lower temperature limit may have to be raised for satisfactory operation at higher temperatures. However, the new generation of catalysts developed specifically for the methanation of coal gasifier effluents are overcoming the earlier deficiencies of the older commercially available catalysts.

For most of the catalysts studied, the hydrogen-to-carbon monoxide ratio in the feed gas should not be less than 2.85 for the temperatures between 525° and 900°F and for pressures above 600 psig. The catalysts that were deactivated by an even lower H<sub>2</sub>:CO ratio and/or undesirable temperatures could not be regenerated.

The presence of large amounts of methane in the feed gas has a small effect on the rate of methanation as long as the relative partial pressure of methane is low, as compared to that of hydrogen. More studies are required in this area, especially for low-pressure coal gasification processes.

The presence of 1 mole percent (or less) benzene does not affect the methanation catalysts. The presence of higher concentrations of benzene tends to reduce methanation activity gradually and in steps. Furthermore, the methanator temperature must not be allowed to increase uncontrollably while benzene is present; otherwise, carbon will be deposited. Most of the catalysts were not regenerable after benzene deactivation.

If both CO and CO<sub>2</sub> are present in the feed, CO<sub>2</sub> will be hydrogenated to methane only when the concentration of CO is less than 0.2 mole percent. Excess CO<sub>2</sub> (15 percent or more) may hinder the CO methanation reaction.

The presence of nitrogen in the coal gasifier effluent (methanator feed) may cause ammonia formation because nickel and iron in the catalyst (or in the

reactor walls) promotes ammonia formation in the absence of carbon oxides. Ammonia tends to deactivate the methanation catalyst, but it can be regenerated by passing hydrogen at about 700°F. However, carbon deposition usually follows deactivation.

The effect of sulfur on the catalyst activity was determined in life-test type runs. Generally, small concentrations (< 1 ppm) of propyl mercaptan and thiophene do not affect the nickel catalysts. However, methyl mercaptan, carbonyl sulfide and hydrogen sulfide in excess of 1.2 ppm quickly poison the nickel catalysts. Carbon deposition took place after deactivation. Neither the sulfided catalysts nor the carbon deposited catalysts were regenerable.

Life testing results of three promising catalysts are summarized.

The existing coal gasification processes produce a reactor output gas (prior to clean-up and methanation) with a heating value of from 300 to 600 Btu/SCF per standard cubic foot; this consists primarily of methane, hydrogen and carbon oxides. In order to produce a gas with a heating value of about 1000 Btu per standard cubic foot and interchangeable with pipeline gas, methanation of the above-mentioned gas is essential. Heterogeneous catalysis — the heart of the methanation process — is by nature difficult and inconsistent, because:

- First, there is no guarantee that two batches of catalysts will be made exactly alike.
- Second, there is no guarantee that the molecules which meet the catalyst will have the same number and the same composition at all times.

The critical aspects of the methanation process are catalyst performance and the removal of the exothermic heat of reaction.

## 1.6 Summary of Coal and Char Characterization

The summaries of sub-parts in this category are presented under individual subheadings because of disparate content.

### 1.6.1. Petrography and Related Studies

Petrographic and related properties of coal were investigated at different stages in the hydrogasification process. Particles of caking bituminous coal inflate to round, hollow "cenospheres" when they are pretreated with air or diluted air, in a fluidized bed, and at elevated temperature, to destroy their tendency to agglomerate. The loss of the agglomeration tendency is attributed to a surface transformation, which forms a "skin" or "envelope" or "oxidized layer" of reacted coal that can be distinguished by its high reflectance. IGT believes this sheath remains rigid, and that this rigidity tends to limit further enlargement of particles during later, high-temperature stages of the HYGAS process. Both the pretreated particles and residue particles from hydrogasification vary greatly in structure and reflectance. The caking mechanism of pretreated low-volatile bituminous coal in the hydrogasification reactor was also investigated.

Petrographic study of the specified feed coals and some of their chars at different stages of processing has been fruitful in several ways. The detection of contaminating caking coal mixed with a nonagglomerating coal explained some anomalous hydrogasification processing results, although the examination was not early enough to avoid their occurrence. Agglomeration of pretreated high-rank bituminous coals was shown to result from the discharge of fluid material from the interior of pretreated particles as well as from the fluidity of the few particles that escaped pretreatment. Lack of anisotropy in the residue chars from lignite, subbituminous coal, and pretreated high-volatile C coal indicates that little or no fluidity developed in these coals, and accounts for the successful processing of lignite and subbituminous coal without pretreatment. Increasing amounts of anisotropy, indicating development of fluidity, were observed in chars from the higher rank bituminous coals. Increased anisotropy and a very different char particle structure were found when a high-volatile A bituminous coal was fed without pretreatment.

Differences in structure may become important if process development makes it possible to feed the coal without pretreatment. Because graphitization at higher temperature occurs only when the char has passed through a fluid stage with resulting anisotropy (Kipling and Shooter<sup>1</sup>), the degree of anisotropy or lack of it may indicate differences in the electrical characteristics of the char; this may affect its behavior in the electrothermal gasification process for the production of hydrogen.

Correlating the petrographic properties of a coal and its suitability for hydrogasification, we find:

- Exinite is largely lost in pretreatment and, therefore, is of value only if the coal can be processed without pretreatment.
- Fusinite and semifusinite are expected to be less reactive than vitrinite but no quantitative data on this reactivity are yet available.

### 1.6.2 Miscellaneous Support Studies

Laboratory tests on the slurring of coal and char were performed prior to the pumping tests. Raw or pretreated Ireland Mine coal was easily wetted and formed into a slurry by stirring with water. Water removal from the slurry by centrifugation was investigated; pressuring the slurry substantially increased the amount of nonrecoverable water. Also investigated were slurring with benzene, the separation of benzene and water in the presence of coal fines, and the use of light oil rather than benzene as the slurry medium — the use of toluene proved to be a feasible alternative. In an investigation of fines formation from sudden heating of the pretreated coal slurry, only a small amount of fines was produced. The float-sink properties of chars were tested at different stages of hydrogasification; results indicate that the float-sink property depends greatly on the amount of noncondensable gas left in the pores of the residues after submersion.

Other support studies included the analyses of condensate water samples for phenol and hydrogen cyanide; analyses of the hydrocarbon liquids from two hydrogasification runs; and the analyses for ammonia in the water phase of the condensate from several runs, in which the results indicated that all or nearly all of the nitrogen was converted to ammonia. Use of the residues from electrogasification for removal of phenols from the water condensate was also investigated.

Minor components in the pilot plant hydrogasification reactor product gas were identified by gas chromatography and mass spectrometry.

The attrition resistance of the chars from hydrogasification and other sources was determined by a test developed for this purpose. The quantity of -200 mesh fines produced in the test was considered significant. Attrition resistance was constant or increased slightly with increase in conversion. This is characteristic of the residue as a whole, rather than of the coarse particles only.

### 1.6.3 Calorimetry

Calorimetry research was required to establish valid data on various heats of reactions upon which to base the design of an efficient coal hydrogasification plant. Data were required for:

- Raw coal in the coal pretreatment process,
- Pretreated coal in the low-temperature gasifier,
- Residue from the low-temperature gasifier after that residue has passed into the high-temperature gasifier, and
- Residue from the high-temperature gasifier.<sup>2, 3</sup>

A thorough literature search revealed that no data had been reported on the heat of reaction of hydrogen and coal; therefore, two calorimeters were designed, constructed and operated by the Institute of Gas Technology to obtain the data required. One calorimeter was designed to measure the heat of reaction, and the other to measure heat capacity by the drop method. The heat-of-reaction calorimeter can be operated at temperatures up to 1500°F and at pressures up to 1500 psia. The drop calorimeter can be operated at atmospheric pressure and temperatures up to 1500°F. Results of the following investigations are reported in this section:

- 1) The heat of reaction of hydrogen with coals and coal chars after various degrees of gasification,
- 2) The heat of reaction of coal pretreatment, and
- 3) The heat capacity of various coals and coal chars

Ash balances were used to put these results on a common basis.

The majority of work involving the heats of reaction of hydrogen and coal was concentrated on the study of a high-volatile content bituminous coal from the Ireland Mine's Pittsburgh No. 8 seam. Samples involved raw coal in a size range of -20 to +325 mesh, pretreated coal, residue representative of that from the low-temperature stage of the hydrogasifier and residue representative of that from the high-temperature stage of the hydrogasifier. Some studies also were made on West Virginia Sewell coal (Sewell No. 1 Mine), West Virginia Block No. 5 coal (Kanawha Mine), Illinois No. 6 coal (Crown Mine), Colorado subbituminous coal and North Dakota lignite.

The average heat of reaction in raw coal gasification was about 1800 Btu/lb coal reacted; 52 weight percent of the organic carbon in the feed was gasified.

Examination of the temperature measurement, the pressure measurement, the temperature distribution in the calorimeter, the total mass balance and the calibration results obtained from the constant-heat-input method and the experimental runs on hydrogen and n-decane reactions indicate that the data reported should not have a deviation greater than 10%.

In determining the heat of reaction the coal sample was stored at 70° to 72°F in the neck of the calorimeter. When the calorimeter reached thermal equilibrium with the heater guard and the pressure was stabilized; the sample was lowered into the calorimeter body. In determining the heat of pretreatment, the coal was kept at 70° to 72°F while the air-filled calorimeter was stabilized at desired conditions. The coal was then dropped into the calorimeter after equilibrium condition had been reached; therefore, the measured heat of pretreatment included 1) the heat required to warm the coal from 70°F to the reaction temperature, and 2) the heat of pyrolysis.

During the tests, little devolatilization was noticed at 700°F, but the coal devolatilized rapidly at 800°F, as shown by the presence of tars. Thus, because a good portion of the coal weight loss at 800°F resulted from devolatilization and not from the oxidation reaction, the heat of reaction of the coal pretreatment was also calculated for 800°F, based on the data obtained at 700°F and the heat capacity information of both reactants and products. One should note that, although these experiments were carried out in a static-bed reactor, the IGT pilot plant pretreatment of coal is carried out in a fluidized-bed reactor. At 700°F, the rate of the devolatilization reaction apparently competes with that of the oxidation reaction. In the static reactor the poor gas-solids contacting favors devolatilization, while the intimate gas-solids contacting in the fluidized-bed reactor favors oxidation. For a fluidized pretreater, therefore, the calculated values of the heat of reaction at 800°F more nearly approach the actual values.

Based on the data obtained from this work and that available in the literature, the heat capacity of coal is believed to be a function of the volatile matter content and the temperature. The change of heat capacity with volatile matter content at a constant temperature is nearly constant for every temperature. The heat capacity and temperature are also nearly constant for every constant volatile matter content within the accuracy of the data.

Utilizing the two calorimeters developed by IGT, mean pyro-heat capacities of coal were determined. Results were obtained for temperatures that ranged to >1300°F for lignite, and to >1500°F for raw coal, pretreated coal, low-temperature residue and high-temperature residue. Plots of these data together with other data from the literature are presented.

Based on the data obtained from the work reported here, together with data available in the literature, a comparison chart was developed. Predicted values are plotted for comparison with the experimental data. The largest deviation between predicted and experimental data is about 10%, while the average deviation is about  $\pm 5\%$ .

It should be noted that the results obtained from this investigation are sufficient for a specific application only and should not be used for general analysis or correlation for all applications. To achieve a generalized correlation suitable for providing design information adaptable to any coal gasification process, further studies will be required.

#### 1.6.4 Kinetics of Coal Gasification

A quantitative mathematical model to describe coal char gasification kinetics was developed based on experimental information obtained over a wide range of conditions. The model is intended for application to two reaction stages:

1. A "high-rate methane formation stage" corresponding to a limited period after a coal char containing reasonable volatile matter is initially exposed to a gasifying medium containing hydrogen. During this short-lived period, a coal char exhibits an exceptionally high, although transient, reactivity for methane formation.
2. A "low-rate gasification stage" corresponding to gasification of a char of relatively low reactivity, which results when reaction in the "rapid-rate" stage is completed.

The correlations corresponding to this model were evaluated on the basis of data obtained at constant environmental conditions with a thermobalance apparatus and a differential fluidized-bed system at 1500° to 1750°F and 1 to 70 atmospheres with a variety of gases and gas mixtures.

Consistencies between predictions of the correlations and results obtained from a variety of other experimental gasification systems employing integral gas-solids contacting are excellent.

#### 1.6.5 References Cited

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## 1.7 Summary of Coal Pretreatment in the 10-Inch PDU

The most reactive part of coal for producing methane is contained in the volatile matter of coal, a fact that favors the use of high-volatile coals to synthesize substitute natural gas (SNG) for pipeline use.

As a result of a literature review and batch pretreatment tests with air and nitrogen, both reported in IGT Research Bulletin No. 39,<sup>\*</sup> it was concluded that oxidation with air or diluted air was the most suitable method of destroying caking properties of bituminous coal with minimum loss of the valuable volatile matter.

A fluidized-bed unit was constructed and several tests were conducted at near-atmospheric pressure with Pittsburgh No. 8 seam high-volatile bituminous coal. Tests also were made with this coal at pressures up to 1055 psig utilizing a high-pressure reactor.

The objectives of the work reported here were:

- Development of a satisfactory, continuous, atmospheric-pressure fluidized-bed pretreatment unit and process,
- Establishment of minimum pretreatment conditions for the complete range of rank of bituminous coal, and
- Preparation of pretreated coal for the 4-inch-diameter hydrogasification process development unit.

The investigation continued over the period from October 1964 to July 1969. A total of about 72 reported runs were made on high-volatile bituminous coals, 2 on a medium-volatile bituminous coal and 6 on low-volatile bituminous coals. Toward the end of the investigation, conditions required for minimum pretreatment of coals ranging in rank from high-volatile C bituminous to low-volatile bituminous were determined.

Conditions necessary to destroy the agglomerating tendency of caking coals with minimal pretreatment using air or diluted air were established in an atmospheric-pressure fluidized-bed reactor. For high-volatile bituminous coals, which are commercially significant for SNG production, the successful pretreatment conditions were:

- 1) Temperature above 735°F,
- 2) Oxygen consumption of 1.0 to 2.5 standard cubic feet per pound of coal fed, and
- 3) Residence time exceeding 10 minutes.

The volatile matter in the coal was reduced from a range of 33 to 39% to a range of 24 to 26% before free-flowing char was produced which could be continuously fed to the hydrogasifier.

\* IGT Research Bulletin No. 39:  
Production of Pipeline Gas by Hydrogasification of Coal, Volume 1,  
1954-1964, IGT and A.G.A., Chicago, December, 1972.

## 1.8 Summary of Commercial Plant Design

The design of a HYGAS demonstration plant is discussed in Part VIII; the design is based on technology existing in December 1969. The design study was undertaken by Procon Incorporated and IGT for the Department of the Interior, Office of Coal Research, and the American Gas Association. The design that evolved could be the basis for one train of the full-scale commercial facility that is illustrated in Figure 1-5.

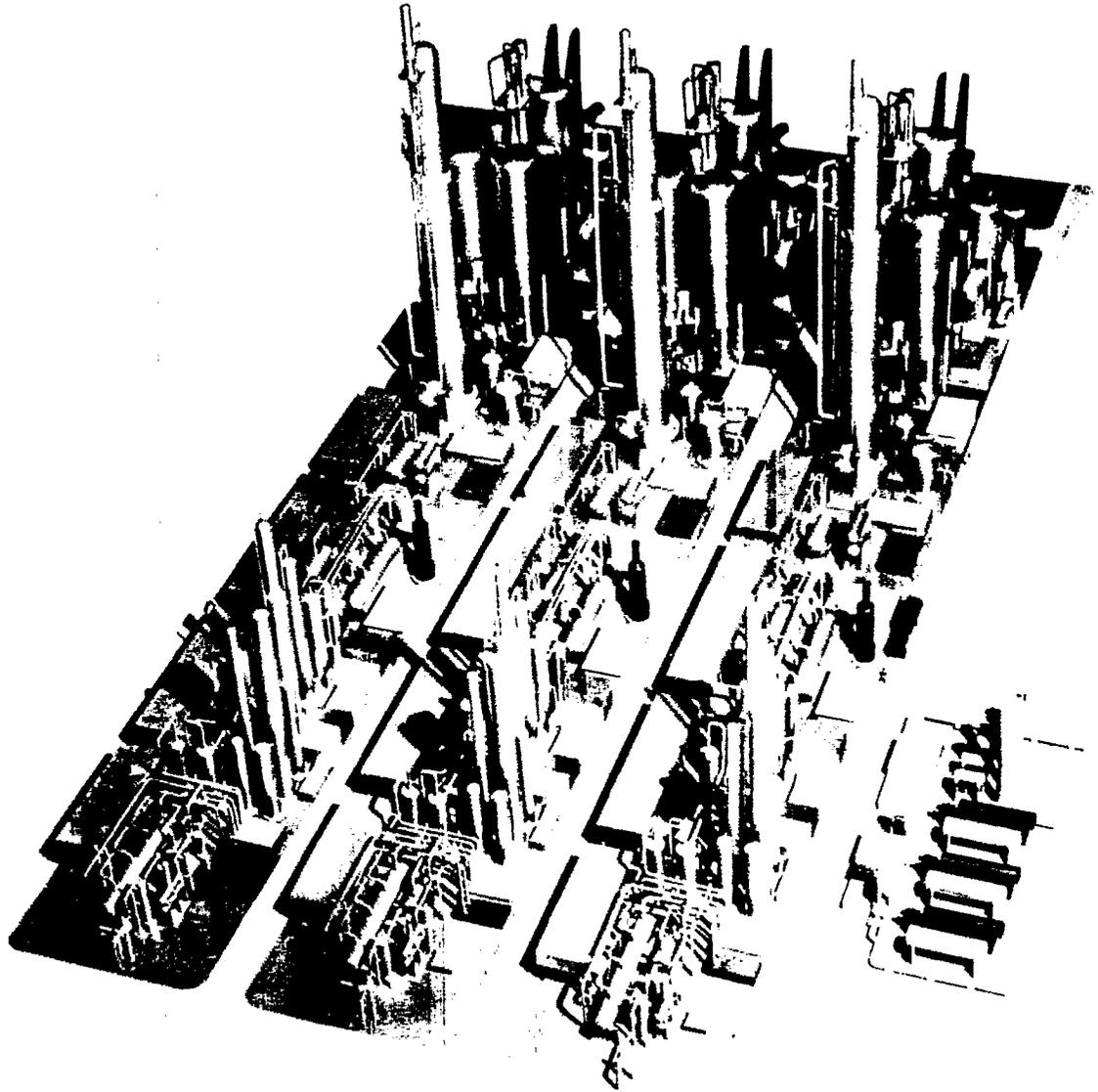
The 1969 plant design incorporated the electrothermal process for manufacture of hydrogen-rich gas because that process was most advanced at that time. Extensive research and development by IGT for OCR and others has since proved that two alternative systems for producing hydrogen-rich gas — namely the steam-oxygen and steam-iron processes — may be even more promising based on current economics. Facilities for extensive testing of these processes at the HYGAS pilot plant are being readied as this report is written.

Because the design discussed was an early design and progress in the hydrogasification of coal to high-Btu gas for pipeline use has been substantial since 1969, a critique of the design also is presented together with suggested areas of improvement.\* Progress has been achieved in most of the improvement areas discussed since this work was performed. The purpose of the early demonstration plant design in 1969 was 1) to provide a guide for future designs, both in the arrangement and sequence of processing steps, 2) to pinpoint problem areas where pilot-scale study should be concentrated, and 3) to pinpoint problem areas where a paucity of existing data might indicate additional basic research would be required; all of these goals have been accomplished.

The primary impact of this design, however, has been to show that it is feasible to design large-scale plants for the hydrogasification of coal to pipeline-quality gas (SNG), with new American approaches to gasification, coupled with existing technology. As any of the desirable improvements discussed herein become available, both the economics and design of coal gasification plants will benefit. This design project indicates, however, that commercial conversion of coal to SNG is feasible today with improved American technology, without awaiting advances to optimum designs.

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\* The Section 8.7 — Suggested Areas for Design Improvement — in this report is based on "Analysis of a HYGAS Coal Gasification Plant Design," presented by F. C. Schora and C. W. Matthews in November 1972 before the 65th Annual Meeting of the A. I. Ch. E. at New York City.



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Figure 1-5. PHOTOGRAPH OF THE MODEL OF A COMPLETE, COMMERCIAL HYGAS PLANT THAT COULD CONVERT 15,000 TO 18,000 TONS OF COAL DAILY INTO 250 MILLION CUBIC FEET OF SNG FOR PIPELINE USE. THE DEMONSTRATION PLANT DISCUSSED IN PART VIII COULD COMPRISE ONE PROCESS TRAIN SIMILAR TO THE THREE THAT COMPRISE THE PLANT SHOWN

## 1.9 Summary of Process Economics

### 1.9.1 Discussion

Several economics studies pertaining to the HYGAS Process conversion of coal to pipeline-quality SNG were performed between July 1964 and September 1972, the period reported in this OCR R&D Report No. 22.

The reader should bear in mind that cost estimates are usually subject to comparison, if not in a formal tabulation, then more casually in the reader's mind with reference to some prior knowledge. One must always keep in mind the comparability of different estimates.

Specifically, one should recognize that process design, degree of detail, source of costs, timing of estimates, cost of coal, financial factors and methods employed to arrive at a process price may vary greatly between analyses.

With these thoughts in mind, the reader is referred to the bibliography that pertains to the economics of producing pipeline-quality SNG from coal; all were prepared by HYGAS engineering economists during the contract period, and explore the economics of the HYGAS Process in that period.

The reader is referred, also, to the Final Report of the Supply Technical Advisory Task Force - Synthetic Gas-Coal by the Federal Power Commission dated April 1973, specifically, Chapter VII, "Economics of Pipeline Gas From Coal," paragraph B, Processes Evaluated *et. seq.* The "single set of data" felt to be representative of a "Typical New Process," although not actually HYGAS data, are, for practical purposes, similar.

### 1.9.2 Bibliography

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## 1.10 High-Btu Bibliography

NOTE: For additional information on this subject, see HYGAS: 1972 to 1974, Pipeline Gas From Coal - Hydrogenation, OCR, R&D Report No. 110, Interim Report No. 1. Prepared by Process Research Division of IGT for OCR, the report covers HYGAS Pilot Plant operations to the end of June 1974, Ash-Agglomerating Gasification Tests, Steam-Oxygen Modifications, Checkout and Shakedown of the 2.25-Megawatt Electrothermal Gasifier System, and various HYGAS Support Studies (1 vol, 220 pp). In preparation, for publication during 1975.

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