

INSTITUTE OF GAS TECHNOLOGY

CONF-760838-2

HYGAS[®] PROCESS UPDATE

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Presented at the

Third Annual International Conference on
Coal Gasification and Liquefaction
University of Pittsburgh
August 3-5, 1976

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INTRODUCTION AND HISTORICAL BACKGROUND

The HYGAS[®] Process involves hydrogen gasification of coal and derives its name from that reaction. The main product of the process is substitute natural gas. The HYGAS Process is a second-generation high-pressure process designed to handle all ranks of coal. It is a method which can produce approximately two-thirds of the product methane in the hydrogasifier by direct hydrogenation. The process is carried out at a pressure of about 1000 psi and temperatures from 600 °F in a slurry drier to 1850 °F in the steam-oxygen gasification zone. It is currently being studied in a pilot plant designed to produce 1-1/2 million SCF/day of methane from 80 tons of coal. This presentation will give some background on the process and an indication of our progress to date.

Early studies of hydrogen gasification of coal at the Institute of Gas Technology (IGT) go back more than two decades. This led to contracts with the U.S. Department of the Interior, Office of Coal Research, and support by the American Gas Association (A.G.A.) starting in 1964. The objectives of the HYGAS program are to verify the technical and economic feasibility of the process and to develop the HYGAS concept as a candidate commercial process by the 1980's, as well as to develop an environmentally acceptable coal conversion process that can utilize all

types of coal. It could supplement natural gas supplies and reserves with gasified coal. This program is one of several pilot plant programs currently being supported by the U.S. Energy Research and Development Administration (ERDA), and in some cases also by the A.G.A. These processes include the Synthane Process, the CO₂ Acceptor, BI-GAS, and the Agglomerating Ash Burner.

THE HYGAS PROCESS

The HYGAS Process is based on two processes for converting coal to synthetic pipeline gas. One of these involves the gasification of coal with oxygen and steam to produce a mixture of carbon monoxide and hydrogen which is suitable for methanation. The other involves direct hydrogenation of coal at high temperatures (1200° -1800°F) and high pressures (1000 psi) at moderate residence times. The present HYGAS Process incorporates principles developed in both of these concepts.

The resulting HYGAS Process gives a high yield (65-70%) of the product methane formed in the hydrogasifier by direct hydrogenation of coal. The use of hydrogen and steam in hydrogasification gives an opportunity to balance exothermic reactions with endothermic reactions in order to provide built-in temperature controls as well as internal hydrogen generation.

The HYGAS Process as presently operated is illustrated in Figure 1. Raw coal is crushed to pass a 1/8-inch screen and, if it possesses agglomerating properties, is sent to a pretreater. Otherwise, this step is omitted. The pretreater is a fluidized bed which permits a surface oxidation of the coal at atmospheric pressure and temperatures

in the vicinity of 800 °F with air. Some fuel gas is released in this step. The pretreated coal goes to slurry preparation which is not shown. This slurry is pumped to the top of the hydrogasifier. In the uppermost stage the light oil is removed and the dry solids contact a hot rising stream of hydrogen and other gases. The partially reacted coal falls to a lower portion of the reactor to have a further opportunity to react at a higher temperature with more hydrogen.

The residual char then passes to a steam-oxygen gasifier where the temperature is increased and oxygen reacts with carbon to form carbon monoxide which in turn may react with steam to form hydrogen. The spent material is discharged to the ash receiver. The product mixture goes to a gas purification train for quenching, removal of the light oil and carbon dioxide as well as sulfur compounds and ammonia. The remaining gas mixture, essentially carbon monoxide and hydrogen besides the methane, is in a desirable proportion for converting to methane in the methanator. The product gas may then be dried and placed in the pipeline.

Figure 2 indicates a cross section of the HYGAS pilot plant reactor. The structure is about 132 feet tall. It has been constructed as a balanced pressure reactor with a maximum 30"-ID for the lower stages. The upper part of the reactor is pressurized between the shell and components with nitrogen. Pressures of 1000 to 1100 psi are maintained in the reactor.

Coal enters at the top as a slurry of up to 50-50 weight percent mixture with a light oil produced in the process. The upward flow of hot

product bases creates a drying zone in which the light oil is removed. The dried coal passes down to a mixing chamber to then undergo first-stage hydrogasification at about 1250 °F. Residence times vary up to 10 seconds before entering the gas-solids disengaging zone. Partially reacted char then goes from the first-stage hydrogasification to the second stage. This is a large fluidized bed where the temperature is increased to about 1750 °F depending on coal characteristics. A residence time of about 30 minutes is provided here. Char from second-stage hydrogasification is discharged to the steam-oxygen gasifier in the lowest part for steam and oxygen to produce synthesis gas for the upper stages. The gas flow proceeds vertically upward through the reactor. The raw gas contains a mixture of methane, carbon monoxide, carbon dioxide, hydrogen, steam, ammonia, sulfur bearing gases, and light oils as well as coal fines which did not react.

The first step in treatment of the raw gas is to quench it with a water spray. This gives a three-phase mixture of light oil, water and coal fines in addition to the product gas. The liquid-solid mixture is separated in a horizontal separator. Light oil is recycled to the slurry mixer. Coal fines are separated, filtered, and removed. The water is processed and recycled.

The product gas then goes to a diglycolamine scrubber to remove hydrogen sulfide and carbon dioxide. The amine goes to a steam stripper for removal of hydrogen sulfide which goes to a Claus plant for conversion to elemental sulfur. The lean amine solution is pumped back to the process gas scrubber.

The processed gas then goes to a two-stage methanation unit for conversion of residual hydrogen and carbon monoxide to methane.

EARLY DEVELOPMENT

The history of the development of the HYGAS Process goes back to the period 1944 to 1964 in which IGT carried out in-house and A.G.A. - funded work on the fundamental investigation of concept parameters. A contract signed in 1964 with OCR and A.G.A. provided joint funding to carry out process development unit studies to form the basis for a pilot plant design. In 1967, the design of the pilot plant was started. This was followed by construction and shakedown, as well as the development of an electrothermal gasifier. In 1971, the pilot plant operations were initiated with hydrogen produced from a reformer and an electrothermal gasifier was constructed. Also a steam-oxygen system for gasifying char was developed. In 1972 through 1974 the pilot plant systems were demonstrated in integrated operation. In 1974 the steam-oxygen gasification system was designed and installed. A light oil recovery unit was installed. Operations with lignite and bituminous coals have been carried out.

In the period 1944 to 1964, during the pregovernment sponsorship era the A.G.A. provided \$3 million to fund efforts in a fundamental investigation of the concept parameters. During this time, synthesis gas was generated by steam-oxygen gasification of coal at rates up to 1500 lb/hr. Total methanation of synthesis gas was accomplished on nickel catalysts. The concept of high pressure (1000 to 1500 psi) and high temperature (1200° to 1800°F) hydrogasification of coal was established with hydrogen-steam mixtures. Hydrogasification tests were conducted in batch and continuous flow units in the range of 5 to 10 lb/hr of coal.

In the period of 1964 to 1967 contract efforts under partial OCR sponsorship led to process development unit (PDU) studies to form the basis for a pilot plant design. This work established the fluidized bed rather than a moving bed as a mode of gas-solids contact for ease of scale-up. Continuous PDU gasification tests were conducted at coal feed rates up to 100 lb/hr in a 4-inch-diameter reactor. The two-stage hydrogasification concept was verified for maximum direct formation of methane from coal over a temperature range up to 2000 °F and pressures up to 2000 psi. Thirteen coals were tested ranging in rank from lignite through semi-anthracite. Pretreatment conditions were defined in a 10-inch-diameter PDU. Methanation catalysts were screened and a cold gas recycle system was developed for temperature control in methanators.

HYGAS PILOT PLANT

The period 1967 through 1971 extended the development, initiated the design, and carried out the construction of the pilot plant. IGT, together with Bechtel, developed a preliminary design of the pilot plant. This included the gasifier reactor design which involves multiple reaction stages in one vessel. The single-vessel concept eliminates thermal expansion problems. The gasifier internal diameter was set at 30 inches to reduce reactor heat loss and provide optimum intermediate scale-up data. Slurry feeding of coal was selected. The unreliable life of lock hopper valves led to design of a slurry system for feeding coal into and discharging residue from the gasifier. The detailed design, procurement, and construction of the pilot plant was carried out by Procon Incorporated and was followed by the shakedown of the pilot plant. The shakedown of the pilot plant was carried out from May 1971 to October 1971. The electrothermal

gasifier was also developed. The plant was designed to convert 80 tons of coal/day to 1.5 million cubic feet of pipeline-quality gas. It was also designed to handle lignite, bituminous, and subbituminous coals.

Construction was carried out under a guaranteed maximum price (GMP) arrangement for a \$7 million total. An electrothermal method of gasification for generating hydrogen from char in a 6-inch-diameter PDU reactor with a power input of 300 kW was tested at 1000 psi pressure and 1700° to 1900°F.

The period 1971 to 1972 included plant operation, construction of the electrothermal gasifier and development of the steam-oxygen gasification method for hydrogen production. In this time, seven initial tests were conducted using hydrogen from the reforming of natural gas. The electrothermal gasification pilot plant unit was designed and constructed by Procon to use up to 2 MW of ac power using on-line computer control for power input. It was also designed for integration with the HYGAS reactor for char feed and constructed under a GMP arrangement for \$2 million. Studies of the steam-oxygen gasification method were completed for hydrogen generation from char in the 6-inch-diameter PDU reactor at pressures up to 1000 psi and temperatures between 1600° and 1900°F.

Demonstration of pilot plant systems took place during 1972 through 1974. The use of coal-oil slurry system was demonstrated for pressurizing coal feed, as well as the operability of IGT's cold gas recycle system for methanation. A refractory grid was developed for gas distribution and fluid bed support. Twenty tests were conducted with Montana

lignite culminating in Test No. 27 in March 1974. The test program was completed on batch gasification of char in the electrothermal gasifier. Operability was demonstrated (10 tests conducted). Further development of the unit was not pursued due to the high cost of conventional power generation.

SIGNIFICANT TESTS

The period of 1974 and 1975 involved design and installation of additional pilot plant equipment as well as operations with lignite and bituminous coal. The steam-oxygen gasification unit was designed, constructed, and integrated into the bottom of the HYGAS reactor. A light oil recovery unit was designed, constructed, and integrated into the pilot plant. Ten tests were conducted using lignite with steam-oxygen gasification. Test No. 37, completed in July 1975, was the most successful test in the pilot plant lignite program. The results of this test were self-sustained operation for 15 days. Over 1000 tons of lignite were processed. During the 15-day period the entire plant was onstream for 92% of the time or 330 hours. Steady-state conditions were achieved for 160 hours. Solids flow through the reactor was smooth. Amine acid-gas removal and methanation sections worked well. Full conversion of carbon monoxide was maintained. The light oil stripping section performed satisfactorily for the first time, and net production of oil was demonstrated. Overall material balances for three major sections, that is gasification, purification, and methanation, were essentially closed. The slurry feeding system operated at feed rates up to 2-1/2 tons/hour. The plant was in excellent condition on post-test examination. The test results were reviewed by the ERDA/A.G.A. Operating Committee and judged to have met the requirements of technical feasibility.

Pilot plant operations with Illinois No. 6 bituminous coal occurred from 1975 to the present. Pilot plant operations started in September 1975 with the bituminous coal. Test No. 54 achieved self-sustained operations for nine days of which seven were at 2-1/2 tons/hour and two were at 3 tons/hour of pretreated char to the reactor. This test gasified 640 tons of high sulfur caking coal. Self-sustained operation means that the plant produced gas from the chemical reaction of water, oxygen, and coal without supplemental sources of hydrogen and heat.

In this run, the coal was pretreated to 800 °F prior to being fed into the reactor vessel. Good heat and material balances were recorded during the 214-hour run. The run was terminated voluntarily when the coal supply was depleted.

SUMMARY

In summary, the overall accomplishments of the HYGAS program to date are that it has demonstrated the key process concepts and integrated unit operations. It has also demonstrated several methods of hydrogen generation, including catalytic steam reforming of natural gas, electro-thermal gasification, and also steam-oxygen gasification. A total of 37 tests were conducted with lignite, including a total of 5500 tons of lignite processed. During this time, it demonstrated the technical feasibility of a process using lignite. A total of 17 tests were conducted involving a total of 3100 tons of bituminous coal. Some specific objectives of the HYGAS program for fiscal 1977 include tests to be conducted with subbituminous coal. Data will be collected for use in the design of an effluent treatment and water reuse cycles in a commercial plant. New methanation catalysts will be tested. Materials testing will continue.

Refractory and metals coupons prepared by separate contractors under sponsorship of the Metal Properties Council will be tested. Data will be gathered on performance of piping and vessels in corrosive or erosive environment.

ACKNOWLEDGMENT

The author wishes to acknowledge the support of this work given by the U. S. Energy Research and Development Administration as well as the American Gas Association. Thanks are given for permission to publish this material.

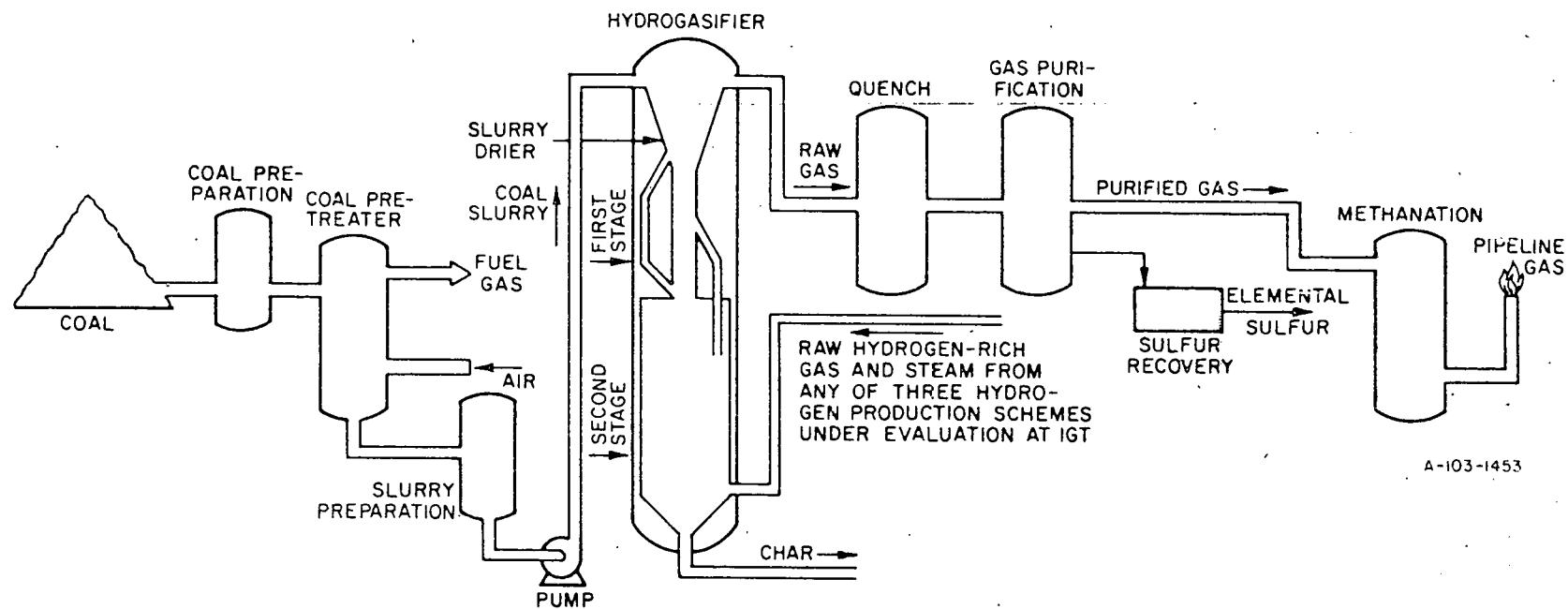
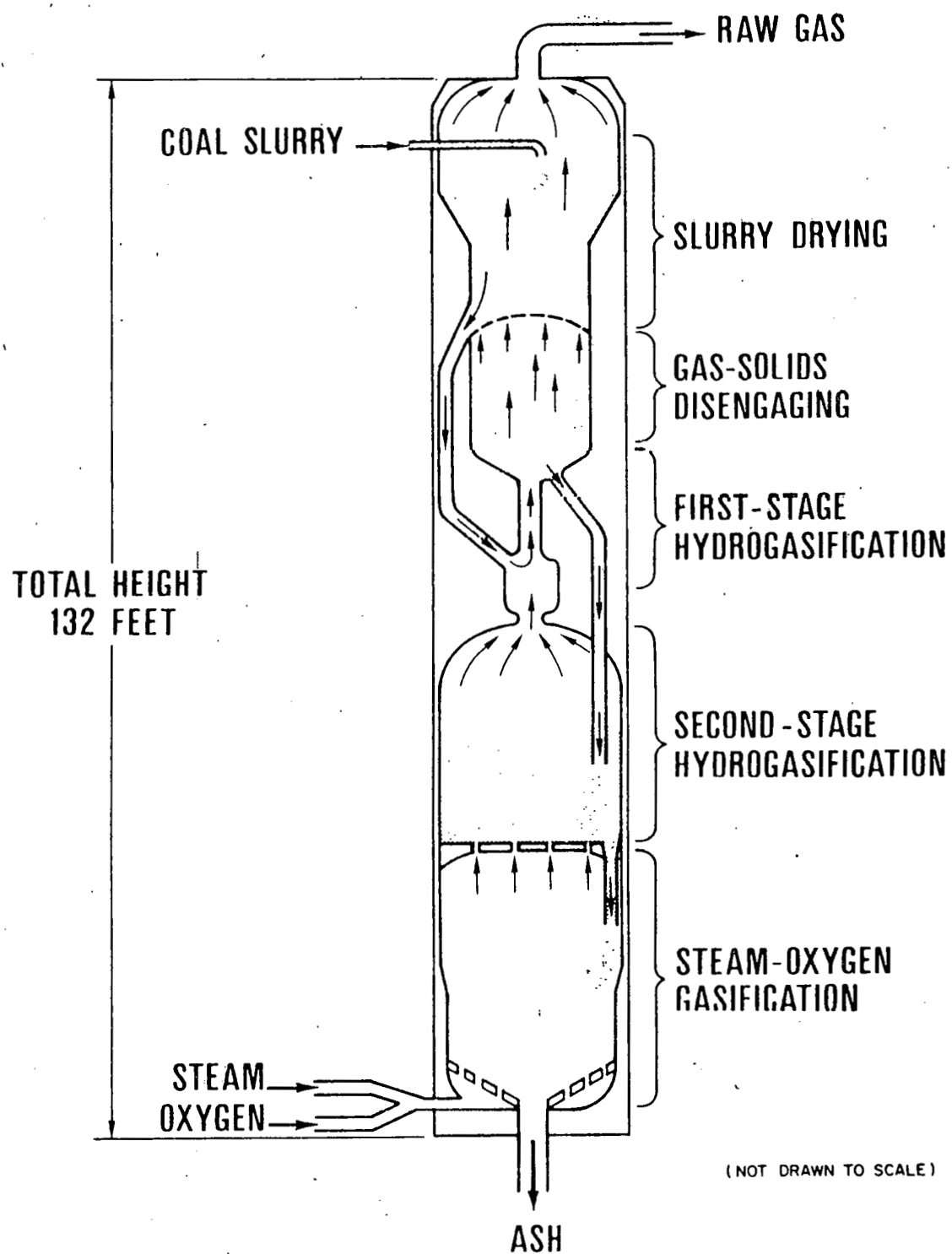


Figure 1. SIMPLIFIED FLOW DIAGRAM OF THE IGT HYGAS PILOT PLANT



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Figure 2. HYGAS PILOT-PLANT REACTOR