

HYDROGEN: A KEY TO THE ECONOMICS OF PIPELINE GAS FROM COAL

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INTRODUCTION

The objective in manufacturing supplemental pipeline gas is to produce high-heating-value gas that is completely interchangeable with natural gas — essentially methane. Large amounts of low-heating-value constituents like hydrogen or carbon monoxide or inert diluents like carbon dioxide or nitrogen cannot be tolerated.

BASIC PROCESS CONSIDERATIONS

The basic problem in making methane from coal is to raise the H_2/C ratio. A typical bituminous coal may contain 75% carbon and 5% hydrogen, a H_2/C mole ratio of 0.4:1; the same ratio for methane is 2:1. To achieve this ratio it is necessary to either add hydrogen or reject carbon. The most efficient way is to add hydrogen. The hydrogen in the coal can supply about 25-30% of the required hydrogen, but the bulk must come by the decomposition of water, the only economical source of the huge quantities needed for supplemental gas.

There are two basic methods for adding hydrogen to coal: In the first, or indirect, method, coal reacts (by Reaction 1) with steam to form synthesis gas — mainly hydrogen and carbon monoxide.



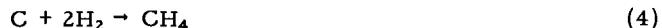
This reaction is highly endothermic and requires combustion of carbon with oxygen, or some other heat source. The CO and H_2 then react catalytically to form methane:



Prior to methanation, part of the CO is made to react with more water to increase the H_2/CO ratio.



In the second, or direct, method, methane is formed directly by the destructive hydrogenation of coal by the reaction:



The indirect method is inherently less efficient because in the process water is decomposed in Reactions 1 and 3. A portion of the hydrogen product is then converted back to water by Reaction 2. Reaction 2 is more exothermic than Reaction 4. Since Reaction 2 is carried out at a much lower temperature than Reaction 1, this heat is not available. Decomposition of an increased amount of water also consumes more energy in the indirect than in the direct method.

The major effort at IGT has been in hydrogasification, now called the HYGAS Process, because of the originally high oxygen consumption and costs of the synthesis-gas methanation route.

PROCESS ECONOMICS

Process economic studies have been carried out in conjunction with the development program at IGT for pipeline gas from coal. A number of different process designs have been prepared in which the price of gas was reduced from the level of \$1.00 to \$0.50/million Btu. The most important effects on the cost of product gas have resulted from the way hydrogen is generated or utilized in the hydrogasifier; hydrogen has been the key factor in reducing the price of gas.

The original studies cover a period of about 10 years and have somewhat different process and cost bases. In this paper the results of seven different pipeline gas plant economic evaluations are compared. An attempt has been made to adjust these to a common and more current basis for capital and operating costs. Coal costs are assumed to be uniform at 16.1¢/million Btu. The plant size is 250 billion Btu of product-gas heating value. The seven studies are -

1. Synthesis-gas methanation
2. Hydrogasification of coal by a hydrogen/char ratio of 300% of stoichiometric
3. Partial hydrogasification with 50% of the stoichiometric hydrogen rate
4. Hydrogasification with steam-hydrogen mixtures
5. Hydrogen by the steam-iron process
6. Hydrogen from synthesis gas generated by electrothermal gasification
7. Hydrogasification with synthesis gas

The data presented in this paper have all been derived from the earlier studies to which the cited references refer. Because of the adjustments in capacity and cost index made to get a better basis for comparison, the costs differ somewhat from the originals. Sulfur by-product credit has not been included because of different sulfur contents for some of the coals used.

Several simple flow diagrams have been prepared to illustrate the different process schemes. Table 1 gives pertinent data; Figures 1 and 2 show the cost of gas in relation to different hydrogen schemes and net production rates. To permit comparison, gas prices shown are based on the same utility-type accounting procedure. The basic assumptions are 1) 20-year straight-line depreciation, 2) 7% return on rate base (end-of-year undepreciated book value plus working capital), 3) 5% interest on debt, 4) 65:35 debt/equity ratio, and 5) 48% Federal income tax. This results in an average annual return on outstanding equity for the cases shown ranging from 9.3 to 9.5%.

Return on equity is calculated as follows: Debt retirement is 5% of the initial debt. Annual depreciation exceeds annual debt retirement by a constant amount, which is called the surplus. This surplus is used to reduce the outstanding equity, which results in a linearly decreasing outstanding equity. To calculate average percent return on equity, the 20-year average net income is divided by the 20-year average outstanding equity.

Interest rates are presently high; even with some reduction in the future, they will probably be higher than 5%. To maintain attractive return on equity at higher interest, the return on rate base will also have to be raised. For a second set of gas prices, we have raised financial factors to a 7.5% interest rate and a 10.1-10.2% average annual return on equity. The income tax rate and debt/equity ratio are as before. This requires a rate of return of 9% on the rate base, from which both debt and equity return are paid.

Table 1. SUMMARY OF PIPELINE GAS FROM COAL PROCESS (250 Billion Btu/Day High-Btu Gas at 1000 psig, 90% Annual Plant Operating Factor, Gas Prices Calculated by A.G.A. Accounting Procedure, Economics of Different Processes Estimated on a Comparable Basis)

Gas Number Process	Partial Hydrogenation, Hydrogen From Spent Char Steam-Hydrogen Gas Mixtures to Hydrogasifier						
	1	2	3	4	5	6	7
Synthesis-gas methanation	916 20,262	934 18,609	939 16,625	947 17,790	941 17,790	941 17,790	937 17,790
Product Gas, Btu/SCF							
Coal, tons/day (Equivalent to 12,400 Btu/lb)							
H ₂ + CO Generation							
Source							
10 SCF/Day	985	470	414	295	295	308	308
Moles/hr	108,406	51,730	45,602	32,495	32,495	33,863	33,863
Oxygen, tons/day	12,500	6,020	5,440	4,109	--	356,000 ^c	356,000 ^c
Purchased Electric Power, kW	--	--	--	--	--	61.7	61.7
Overall Efficiency, %	49.9	54.3	61	66.2	72.6	65.8	65.8
By-products ^a							
Char Plus Fines, tons/day	--	--	--	1,809	468	4,161	4,557
Low-Btu Gas, 10 ⁶ Btu/day	--	--	--	--	61	--	--
Capital Investment	200	218	186	136	99	98	87
Installed Equipment Cost, \$10 ⁶	235-241	256-262	218-223	161-165	118-121	118-121	105-108
Total Investment Cost, \$10 ⁶ ^b							
20-yr Avg Price of Gas	87.0	89.3	75.8	65.1	54.7	61.7	56.1
Standard Factors, #/10 ⁶ Btu	91.2	93.8	79.7	68.0	56.9	63.9	58.0
New Factors, #/10 ⁶ Btu							

^a Sulfur not included because of different costs.
^b Includes working capital, which increases with higher gas price due to new financial factors.
^c In calculating efficiency, fuel to generate purchased power subtracted from by-product char.
^d 258 X 10⁶ Btu/day.

Depending on the investment level, the effect of the higher financial factors is to raise gas price from 1.9¢ to 4.5¢/million Btu for the investment range covered.

Indirect Methanation - Synthesis-Gas Methanation (Case 1)

The first process, methanation of synthesis gas generated by Texaco steam-oxygen suspension gasification of coal,¹ is shown in Figure 3. Gas made this way is expensive because of the high oxygen requirement and the low thermal efficiency. For a 250 billion Btu/day plant, 12,500 tons/day of oxygen are needed for generation of 985 million SCF/day of hydrogen equivalent (CO + H₂). Investment is \$240 million; product gas costs approximately 90¢/million Btu, depending on financial factors.

Direct Hydrogenation

The rest of the studies are based on the direct hydrogenation of coal char to methane discussed above. They represent a historical and process economic study of major steps in hydrogen usage that have occurred in the development of the HYGAS Process.

Use of Excess Hydrogen (Case 2)

The first economic evaluation for hydrogasification was based on pilot plant data in which a large excess of hydrogen - 300% of the stoichiometric hydrogen/char ratio - is fed to the hydrogasifier² in a fluidized-bed reactor (Figure 4). Nearly complete gasification is achieved. A separate coal stream flows to the gasifier where synthesis gas for hydrogen production is generated. The coal pretreatment step, a low-temperature carbonization process, is more severe than the simpler air oxidation used in IGT's later work. More hydrogen and other volatile matter is lost in the low-temperature carbonization, requiring more net hydrogen input.

With excess hydrogen, the hydrogasifier effluent contains CH₄/H₂ in a 0.32:1 ratio, which is upgraded to a ratio of 8.7:1 by low-temperature separation. This processing step contributes about 15¢/million Btu to the price of gas. Gas price and investment are slightly higher than for synthesis-gas methanation, even though the overall efficiency is higher, because of the higher investment. Even though the net hydrogen rate is less than half that for synthesis-gas methanation, thus cutting oxygen consumption in half, the large excess of hydrogen used in the hydrogasifier requires a compensating expense in cryogenic separation and purification.

Partial Hydrogasification With Less Than Stoichiometric Hydrogen (Case 3)

Further development of hydrogasification showed that it is advantageous to hydrogasify only the more reactive fractions of the coal and to use the less reactive residual char for hydrogen manufacture. By the use of a moving bed, a solids down-flow-gas upflow reactor, and a hydrogen/char ratio only 50% of the stoichiometric, a high-Btu gas is produced in the hydrogasifier.³ In Case 3 the hydrogasifier temperature ranged from 1350° F at the top of the bed to 1600° F at the bottom. The same char pretreatment method was used. A lower temperature and a reduced hydrogen/char feed ratio result in a high-Btu gas, eliminating the need for low-temperature separation. Partial conversion of the char reduces the net hydrogen input because more coal must pass through the reactor, yielding more volatile matter. Compared to Case 2 the investment is reduced 15% and the efficiency is raised to 60%. Savings in equipment and higher efficiency combine to lower gas price by 13¢-14¢/million Btu.

Figure 5 gives a general flow sheet for pipeline gas by partial hydrogasification with spent hydrogasifier char as the basis for hydrogen manufacture. Steam is needed in all cases, but alternative methods employ air, oxygen, or electricity as a basic input.

Figure 6 gives the basic scheme for hydrogen generation by the Texaco-type steam oxygen suspension gasification of spent char. This method is used in four (Cases 1-4) of the seven process economic studies. In all cases costs are based on the system used in Reference 5. As discussed below, electricity can also be used as a heat source. To avoid a repetitive flow sheet, both oxygen and electricity are shown as alternatives; however, the use of electricity is not a part of the Texaco Process.

Hydrogasification With Steam-Hydrogen Mixture (Figures 5 and 6)

An important process and economic development was the successful use of steam in the hydrogasifier. In the current concept, steam and hydrogen in approximately equal amounts are fed to a high-temperature fluidized bed where the above reactions (1, 3, and 4) occur. Since the steam-carbon reaction (1) is strongly endothermic and the hydrogen-carbon reaction (4) strongly exothermic, heat effects tend to balance, and there is not the problem of heat removal that exists when only Reaction 4 occurs. Steam acts as a moderator since, as the temperature rises because of Reaction 4, the rate of Reaction 1 increases. Steam decomposition generates hydrogen in situ, thus reducing the size of the hydrogen section and lowering the price of gas. The hydrogen feed/char ratio is reduced to about 33% of the stoichiometric value. When steam is used, the hydrogasifier effluent contains more carbon monoxide and requires more subsequent methanation than when hydrogen alone is used. About two-thirds of the total methane is made in the hydrogasifier compared to over 90% for Cases 2 and 3. However, the cost of increased methanation is more than compensated for by the other cost reductions resulting from the use of steam.

As shown in Table 1, four of the processes utilize steam with the hydrogen-rich gas. In all these cases the hydrogasifier consists of two stages: a low-temperature first stage of 1300°-1500°F to obtain a high methane yield from the volatile matter in the coal and a high-temperature fluidized-bed second stage of 1700°-1800°F to produce methane and effect the steam-coal reaction. All four of the process designs are based on the same coal rate, coal preparation, and hydrogasification steps derived from the design in Reference 6. Major differences are in the hydrogen section.

The economic effect of introducing steam into the hydrogasifier is shown by Cases 3 and 4: Investment is lowered by 25%. In both cases hydrogen is derived from synthesis gas made by Texaco-type steam-oxygen gasification of spent char. When part of the hydrogen is made in the hydrogasifier, the price of gas is shown to be reduced by 10¢-11¢/million Btu; net hydrogen is reduced by 30%. Case 4 is derived from Reference 5 with modifications, as discussed above, based on Reference 6. The 10¢ differential is confirmed by other studies.

Hydrogen by the Steam-Iron Process (Case 5) (Fuel Gas Associates)

The expense of using oxygen to make hydrogen has stimulated interest in alternative methods. The continuous steam-iron process, shown in Figure 7, offers potential for significant cost reduction. It involved the transfer of the oxygen in water to a stream of iron plus reduced iron oxide that flows between oxidizer and reductor. A stream of hydrogen and unreacted steam flows from the oxidizer directly to the hydrogasifier. Spent hydrogasifier char reacts with steam and air to make a producer gas that regenerates the iron oxide. Since this gas is not part of the product, air can replace oxygen. Power for air compression and other plant requirements is provided by an expansion turbine powered by spent reductor gas. Savings in investment contribute most to the 10¢ reduction in gas price from 65¢ to 55¢/million Btu. The hydrogen rate is the same, but the costs of hydrogen and onsite power generation are greatly reduced. As part of the pipeline gas from coal plant, hydrogen by the steam-iron process costs about 20¢/1000 CF compared to 29¢ for hydrogen by steam-oxygen gasification.

Hydrogen by the Electrothermal Process (Case 6)

Another alternative to steam-oxygen gasification is the electrothermal process (Figure 6). Here resistance heating of a fluidized bed of char operating at 1800° - 1900°F supplies the heat for the steam-carbon reaction, and the steam serves both as a reactant and a fluidizing medium. Compression of high-purity oxygen is eliminated, and the reducing gas is not diluted by CO₂ from combustion. Power must be relatively low cost. Our economics are based on a purchased power cost of 3 mills/kWhr. There is enough spent char to supply needed electricity by either a magnetohydrodynamic or a conventional steam turbine system. Such a system would be adjacent to and integrated with the pipeline gas plant and could benefit from the use of hot char transferred directly as fuel to a fluidized boiler. Hydrogen by this method costs more than by the steam-iron process. The price of pipeline gas is very sensitive to the cost of power. A change of 1 mill/kWhr will change the gas price by 3.3¢/million Btu.

Hydrogasification With Synthesis Gas⁴ (Case 7)

Feeding raw, hot synthesis gas instead of hydrogen can substantially reduce the price of pipeline gas. We have shown the economic effect as applied to the electrothermal process (Figure 8). The synthesis gas is essentially CO and H₂. As H₂ is consumed in the hydrogasifier, CO reacts with the steam present to form more H₂. Because of the lower hydrogen partial pressure, a larger reactor column is needed, but its cost is largely balanced by the elimination of the hydrogen preheat system necessary when cold hydrogen is used. Major cost reductions are in the elimination of the CO shift and purification sections needed to make high-purity hydrogen and in savings in offsite equipment. Gas price is reduced by 5.5¢-6¢/million Btu.

SUMMARY

Important process changes have occurred in the development of the HYGAS Process, resulting in much improved economics. The investment for a 250 billion Btu/day plant has been reduced from over \$250 million to \$120 million. Plant efficiency has risen from 50% to 70%. When computed on a comparable basis, these changes have resulted in reductions in the price of gas from approximately 90¢ to 55¢/million Btu. These process changes are summarized as follows:

<u>Process Change</u>	<u>Price Reduction, ¢/10⁶ Btu</u>
Partial Hydrogasification With 50% vs. 300% of Stoichiometric H ₂ /Char Ratio (Case 3)	14
Use of Steam in the Hydrogasifier (Case 4)	10-11
Use of Steam-Iron Process for H ₂ (Case 5)	10-11
Hydrogasification With Electrothermally Generated Synthesis Gas	9-10

Hydrogasification with electrothermally generated synthesis gas and 0.3¢/kWhr power (Case 6) reduces pipeline-gas price by 9¢-10¢/million Btu from Case 4, with synthesis gas instead of hydrogen accounting for about 5.5¢-6¢. Gas price is then about the same as with hydrogen by the steam-iron process.

The basic IGT scheme as presently conceived consists of three stages of coal conversion as shown in Figure 8: 1) a low-temperature (1300° - 1500°F) first hydrogenation stage, either free fall or upflow, for conversion of the volatile matter; 2) a fluidized-bed second hydrogenation stage where steam and synthesis gas react at 1700° - 1850°F to produce methane, CO, and H₂; and 3) a third-stage fluidized-bed

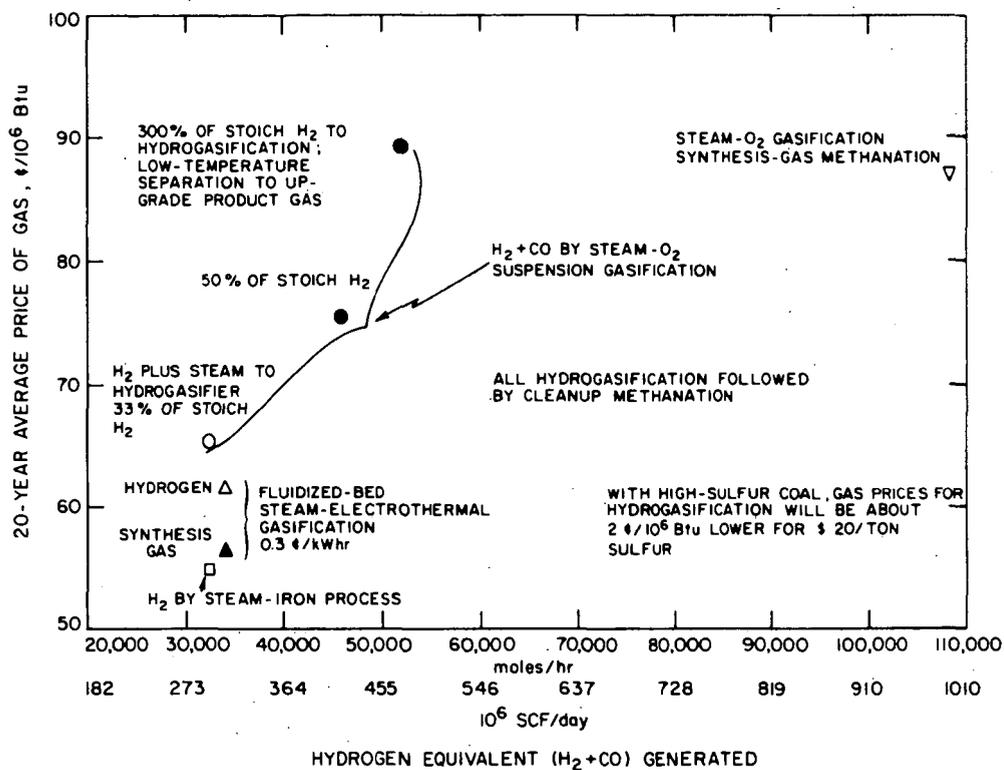
gasifier at 1800°-1900°F where spent char is converted to synthesis gas containing methane by electricity and/or oxygen.

ACKNOWLEDGMENT

The work reported herein is under the cosponsorship of the American Gas Association and the U.S. Department of the Interior, Office of Coal Research. Permission of the sponsors to publish this paper is greatly appreciated.

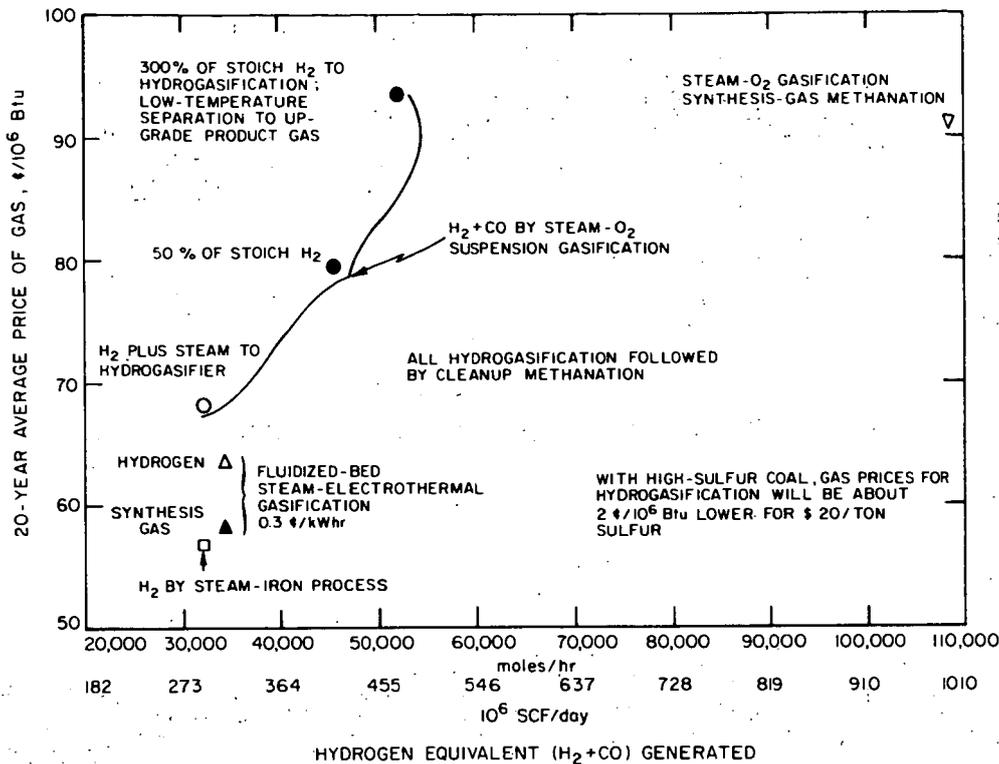
REFERENCES CITED

1. Heffner, W. H. et al., "Pipeline Gas From Bituminous Coal via Suspension Gasification and Catalytic Synthesis of Methane," Study CE-60-207. New York: M. W. Kellogg Co., 1960 (Unpublished report to American Gas Association).
2. Heffner, W. H. et al., "Pipeline Gas From Bituminous Coal via Hydrogasification," Study CE-61-215. New York: M. W. Kellogg Co., 1961 (Unpublished report to American Gas Association).
3. Khan, A. R., Feldkirchner, H. L. and Joyce, T. J., "Techno-Economic Evaluations of Hydrogasification of Coal and Char." Chicago: Institute of Gas Technology, 1964 (Unpublished report to American Gas Association).
4. Knabel, S. J. and Tsaros, C. L., "Process Design and Cost Estimate for a 258 Billion Btu/Day Pipeline Gas Plant - Hydrogasification Using Synthesis Gas Generated by Electrothermal Gasification of Spent Char," R&D Rept. No. 22, Interim Rept. No. 3. Washington, D.C.: Office of Coal Research, 1967.
5. Tsaros, C. L., Knabel, S. J. and Sheridan, L. A., "Process Design and Cost Estimate for Production of 265 Million SCF/Day of Pipeline Gas by the Hydrogasification of Bituminous Coal," R&D Rept. No. 22, Interim Rept. No. 1. Washington, D.C.: Office of Coal Research, 1965.
6. Tsaros, C. L., Knabel, S. J. and Sheridan, L. A., "Process Design and Cost Estimate for Production of 266 Million SCF/Day of Pipeline Gas by the Hydrogasification of Bituminous Coal - Hydrogen by the Steam-Iron Process," R&D Rept. No. 22, Interim Rept. No. 2. Washington, D.C.: Office of Coal Research, 1966.



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Figure 1. EFFECT OF H₂ PLUS CO GENERATION RATE AND METHOD ON PRICE OF PIPELINE GAS FROM COAL ESTIMATED ON A COMPARABLE BASIS (Initial Debt - 65%, Interest at 5%, Return on Rate Base - 7%, Federal Income Tax - 48%, Coal Cost - 16.1¢/10⁶ Btu)



A-70638

Figure 2. EFFECT OF H₂ PLUS CO GENERATION RATE AND METHOD ON PRICE OF PIPELINE GAS FROM COAL ON A COMPARABLE BASIS (Initial Debt - 65%, Interest at 7.5%, Return on Rate Base - 9%, Federal Income Tax - 48%, Coal Cost - 16.1¢/10⁶ Btu)

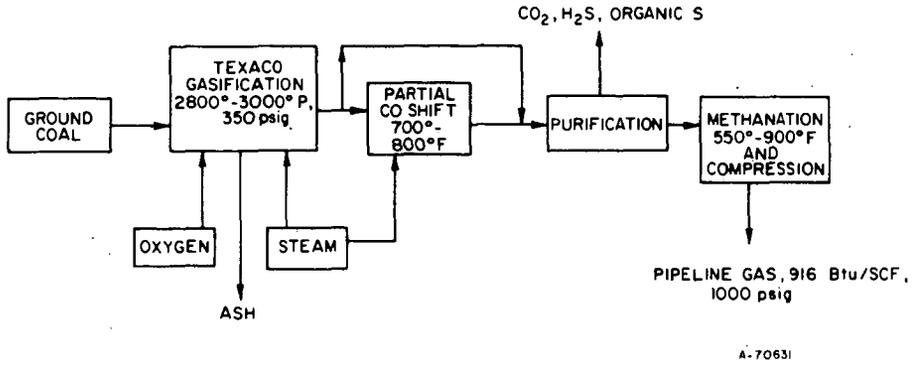


Figure 3. PIPELINE GAS FROM COAL BY METHANATION OF SYNTHESIS GAS

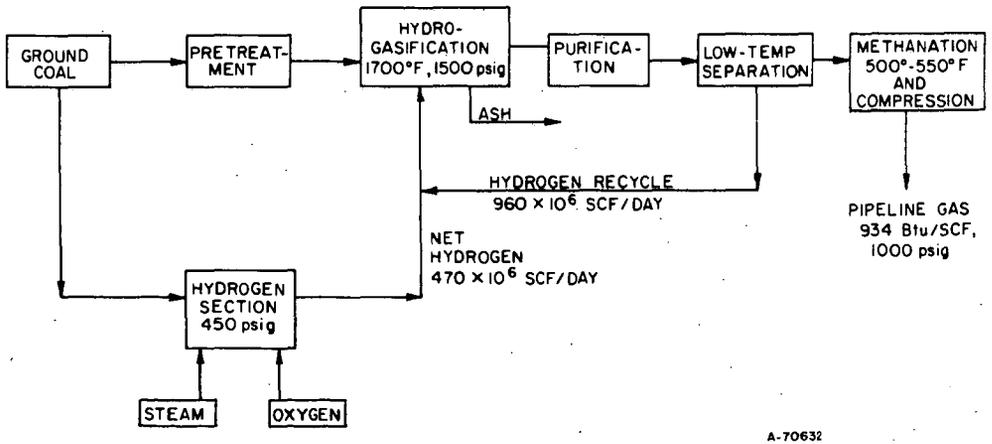
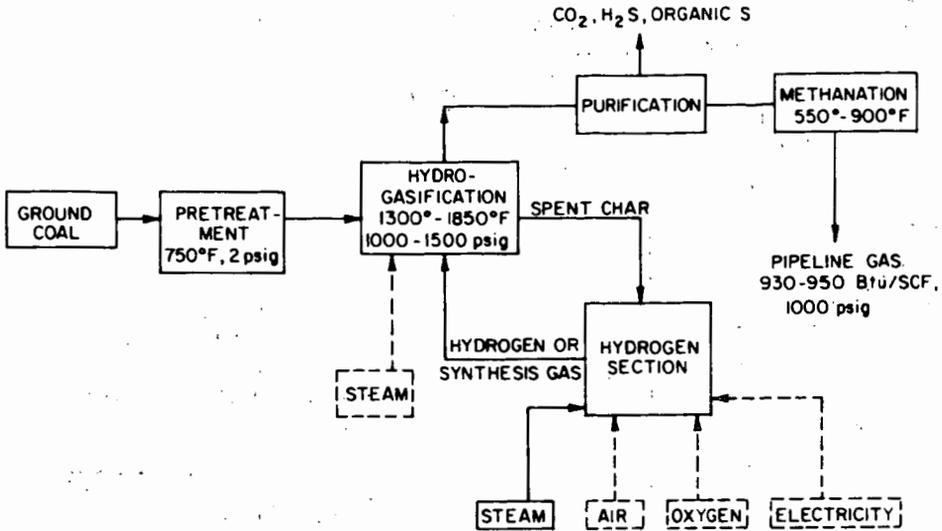
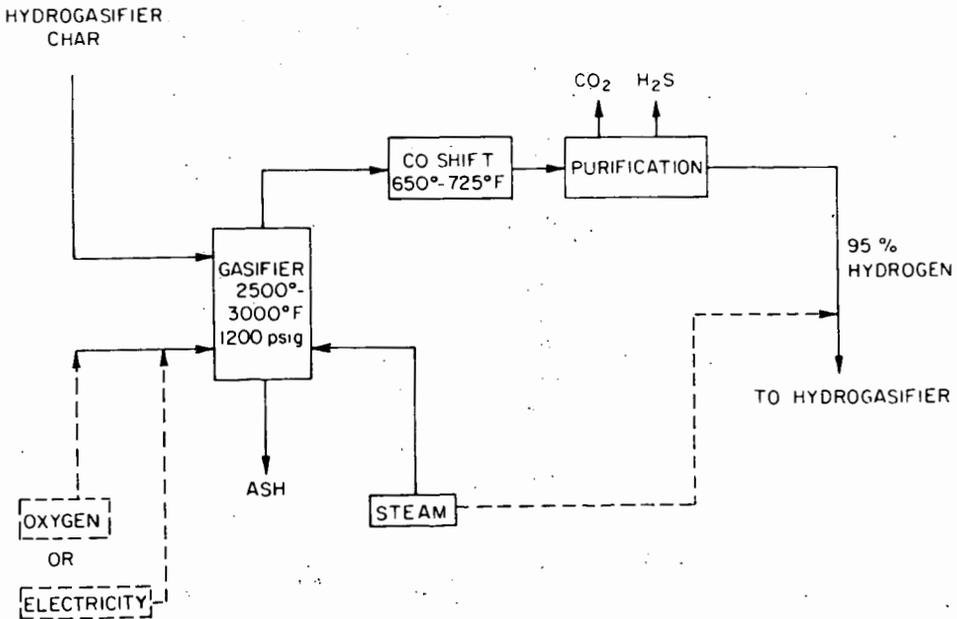


Figure 4. PIPELINE GAS FROM COAL BY HYDROGASIFICATION WITH EXCESS HYDROGEN (300% of Stoichiometric)



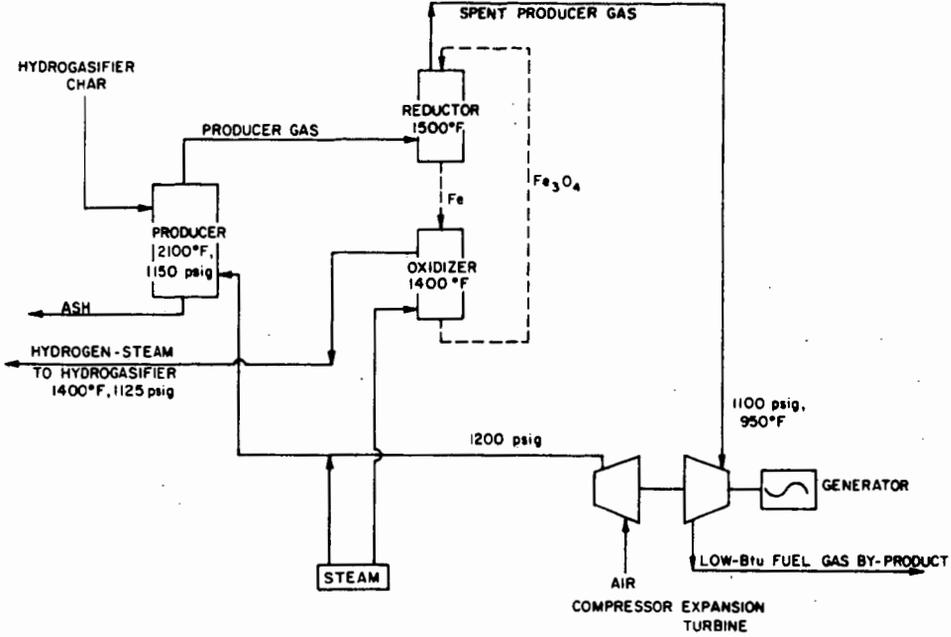
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Figure 5. PIPELINE GAS FROM COAL BY HYDROGASIFICATION WITH LESS THAN STOICHIOMETRIC HYDROGEN; HYDROGEN MANUFACTURE BASED ON SPENT CHAR WITH AIR, OXYGEN, AND ELECTRICITY AS ALTERNATIVES



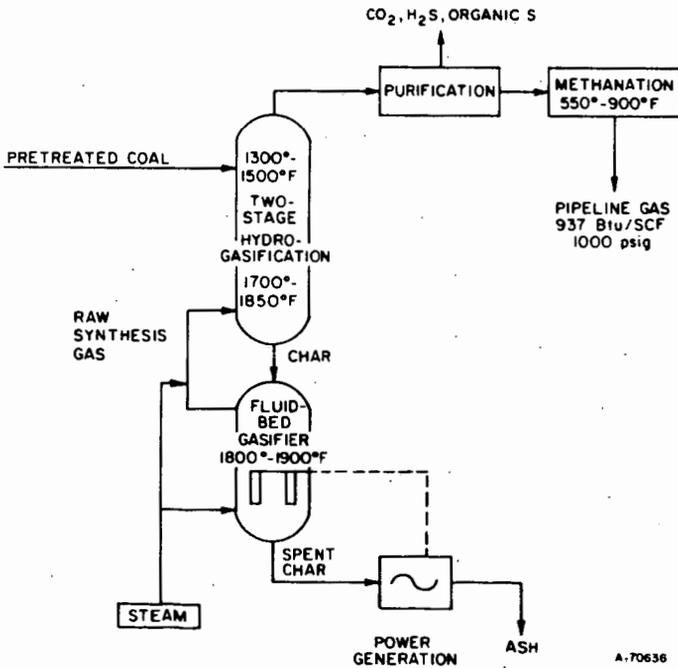
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Figure 6. HYDROGEN GENERATION BY STEAM-OXYGEN OR STEAM-ELECTROTHERMAL GASIFICATION OF HYDROGASIFIER CHAR



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Figure 7. HYDROGEN GENERATION BY THE STEAM-IRON PROCESS BASED ON HYDROGASIFIER CHAR



A-70636

Figure 8. HYDROGASIFICATION WITH SYNTHESIS GAS