# FUEL OIL SUBSTITUTION BY ENRICHED MEDIUM BTU GAS PRODUCED FROM COAL IN WINKLER FLUIDIZED GASIFIERS

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## Abstract

Various carbonaceous materials of carbon contents up to 85 wt% and a wide range of ash types and with levels up to 50 wt% in the feedstock have been successfully gasified in commercial scale Winkler gasifiers to produce -

- 120 Btu/Scf low Btu gas with air blown unit
- 200-300 Btu/Scf medium Btu gas with oxygen blown unit

In order to achieve higher heating value any of the commercially proven coal gasification technologies require additional methane enrichment.

A new development in methanation catalyst technology will allow partial methanation of the gas produced from oxygen blown gasifiers. The methane content in this gas can be increased to produce a gas with a heating value in excess of 400 Btu/Scf.

A methane-enriched medium Btu gas is possible with lower capital and operating costs than those required to produce SNG.

## Coal Based Fuel Gas Production

The production of gases from coal for chemical synthesis and fuel has been well known for many numbers of years and is still used in some parts of the world where coal is in abundance. However, in general, the production of gases from coal has lately become very important due to price increases of oil and natural gas.

The fuel gas from coal based plants has a heating value between 100-300 Btu/Scf, depending upon the oxidizing medium used during the gasification process, and is normally consumed by users located nearby the gasification facilities. As coke oven plants were built, gases with heating values above 400 Btu/Scf became available in abundance and were distributed by pipelines to various chemical industries and to households as "Town-gas". With natural gas availability increasing in the early forties, better gas distribution networks were developed and the use of natural gas increased.

Production of gas as SNG from coal is the most expensive route to generate a fuel gas from coal and is the least thermally efficient.[1] Hence there is increasing interest in making a coal generated gas with a modified heating value of 400 Btu/Scf. The production of such a gas is possible with a conventional proven Winkler fluid-bed gasification and a methane enrichment unit containing a single stage methanator.

### Comparison of Heating Value of Gases

In general, more meaningful comparison of various fuel gases can be seen using the "lower heating values" or better still, the useful heat available above 300°F, (the stack gas exit temperature). The "Higher Heating Value" (HHV) gives natural gas a more attractive image than it deserves. On the other hand, CO and gases containing CO, may not be fully valued if judged by comparing their HHV's with the HHV of methane. The following table illustrates:[1]

Table 1 - SNG Versus Gases Containing CO

| | | HHV | LHV | Useful Heat Above 300°F | Useful Heat % of HHV |
|---|---|---|---|---|---|
| | | Btu/Scf | | | |
| 1 | $CH_4$ (SNG) | 1010 | 909 | 861 | 85 |
| 2 | $H_2$ | 324 | 274 | 261 | 81 |
| 3 | CO | 321 | 321 | 307 | 96 |
| 4 | Particulate free Winkler Gas | 279 | 253 | 238 | 85 |
| 5 | Methane-Enriched Winkler Gas | 450 | 439 | 396 | 88 |

The above table shows that SNG yields 85% of the HHV as useful heat, whereas CO yields 96%, the particulate free Winkler gas 85% and the methane enriched gas 88%.

Many steam producers are contemplating converting their existing natural gas or oil fired boilers to use coal. This conversion to coal is often not practical considering the firing arrangements and configurations for coal. Scrapping the existing boiler and constructing a new coal fired boiler and stack gas clean up is an expensive solution. A more economical solution is to generate a medium-Btu gas from coal in a gasifier and use as it is, or methane-enrich this gas and fire it in the existing oil or natural gas fired boilers.

Although these gases have heating values less than methane, they also require less air for combustion and therefore produce less combustion products than methane. The flue gas ducts, air fans, stack fans and stacks are not limiting factors. Further these gases will combust more rapidly than methane. Additionally, the theoretical flame temperature that can be obtained (over 3500°F) is the same as that from methane combustion, or higher. Table 2 compares the combustion calculations for generation of one million Btu gas above 300°F using theoretical air quantities.

Table 2 - Combustion Comparison

| | Theoretical Air | Fuel Gas | | Flue Gas | |
|---|---|---|---|---|---|
| | LB Mol | LB Mol | LB | LB Mol | LB |
| Methane | 29 | 3.06 | 49 | 32 | 891 |
| Particulate Free Desulfurized Gas Winkler | 18 | 11.07 | 218 | 30 | 890 |
| Methane-Enriched Winkler Gas | 25 | 6.65 | 95 | 31 | 856 |

For a given duty of the boiler, the methane-enriched gas mass rate is twice that of the methane and the corresponding 250 Btu/Scf medium Btu gas is about five times that of the methane.

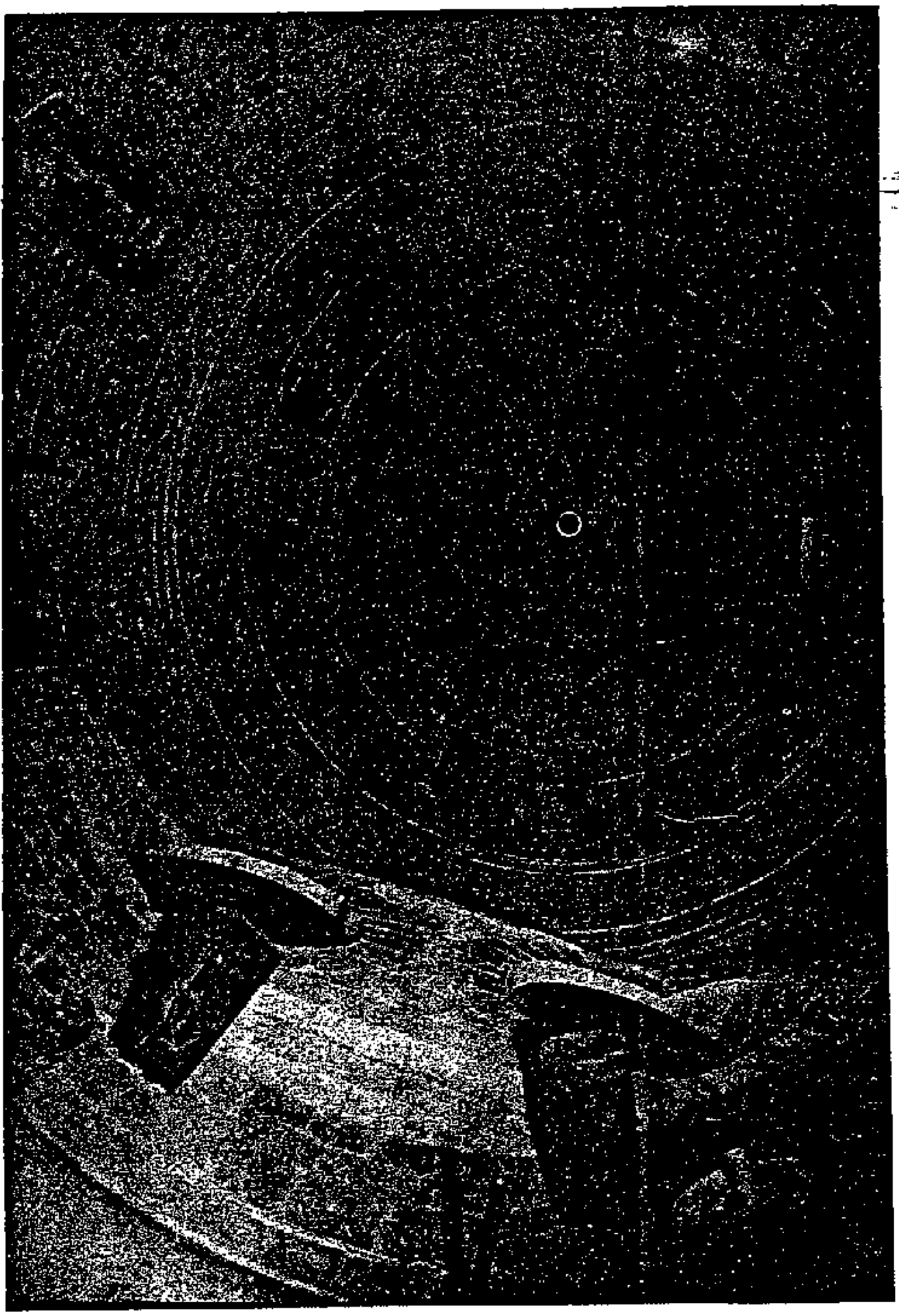
## Fluid-Bed Winkler Coal Gasification Process

The Winkler coal gasifier was the first industrial application of fluid-bed technology. The system was based on the 1922 patent (German Patent No. 437970) awarded to Dr. Winkler for the gasification of coal fines. The first commercial unit put in operation was at Leuna in Germany for the generation of fuel gas to drive gas-machines.[2] This unit was an air blown gasifier producing gases with a heating value of 125 Btu/Scf. The gasifier was 16 ft. (i.d.) diameter and 60 ft. high producing above 3.73 MM SCFH of raw gas. A similar unit, when oxygen-blown, would produce 2.24 MMSCFH of raw gas with a heating value of 270 Btu/Scf. When partially desulfurized and $CO_2$ removed to 1 mole percent (by the BASF Alkazid Process), the fuel gas heating value was upgraded to 320 Btu/Scf. The composition of the raw gas produced from the lignite coal at the plant was:

| Feedstock | Moisture | Ash | C | H | O | N | S | HHV, Btu/LB |
|---|---|---|---|---|---|---|---|---|
| Lignite, Wt% | 8.00 | 12.7 | 56.3 | 4.3 | 15.0 | 0.7 | 3.0 | 9500 |

Gas Produced

| Composition, Mol% | $H_2S$ | $CO_2$ | CO | $H_2$ | $CH_4$ | $N_2$ | HHV, Btu/LB |
|---|---|---|---|---|---|---|---|
| L Btu Gas | 0.8 | 7.7 | 22.5 | 12.6 | 0.7 | 55.7 | 125 |
| MBTU Gas | 1.6 | 17.5 | 41.8 | 37.2 | 0.9 | 1.0 | 267 |
| MBTU Gas ($CO_2$ Removed) | 0.1 | 1.0 | 51.3 | 45.3 | 1.1 | 1.2 | 320 |



View into the conical bottom (fluid-bed zone) of a 10 ft.i.d. Winkler generator with nozzles for steam and oxygen injection, and ash discharge.

- FIGURE 1 -

With the many years of operation, the process has been improved step by step and so much simplified in operation and maintenance that the Winkler Process is known as the most reliable coal gasification process.

The first generation gasifier was initially designed with a mechanically swept, water cooled grid below the fluid bed area. This configuration resulted in gas channelling and hot spots resulting in clinker formation similar to that observed in moving bed or fixed bed type gasifiers.[3]

In the second generation gasifier, this configuration was replaced with a refractory cone with fluidizing gases injected through circumferential nozzles at various levels in the cone.

Figure 1 shows the gasifier with the cone bottom which overcame the problem of clinker formation. Further this resulted in the production of higher CO & $H_2$ in the raw gas, greater once-through carbon conversion and the ability to economically gasify less reactive coals. Additional gasifying media are injected above the fluid bed to further react unconverted carbon in the entrained solids. A radiant boiler (German Patent No. 1421 655) installed in the upper portion of the gasifier prevents any fused ash particles from leaving with the raw gas.

The third generation (described in U.S. Patent No. 4017272) has added pressurized gasification to the other developments in order to increase the production capacity per gasifier for modern mega-methanol plants.[4] A four atmosphere unit is currently offered by Davy McKee. Pressurized gasification reduces capital investment by reducing the number of gasifiers of a given capacity of gas requirement. The four atmosphere unit can produce 6.00 MMSCFH of gas when air blown and 4.00 MMSCFH with oxygen-blown.[5] This quantity of fuel gas, produced in one fluid bed Winkler unit, would allow to substitute approximately 25 ton of fuel oil hourly.

The simplicity and reliability of the Winkler fluid bed operation has been widely demonstrated in about 70 commercial gasifiers. The inherent safety of this process by the large inventory of carbon in the gasifier provides a key factor for consideration, particularly in the oxygen blown gasifiers. Further the process has the advantage that it can handle coal fines from 0-3/8" sizes, which are excessively produced from highly mechanized coal mining operations.[2] Also, the ash content is not critical in Winkler gasifiers. Low grade coals with ash contents up to 50% have been successfully gasified for many years with high on-stream factors of more than 9.90.[6]

## Production of Fuel Gas With a Heating Value of 450 BTU/SCF

The production of a 450 Btu/Scf gas is described herein. The process starts from the conventional, commercially proven Winkler gasification process with a methane enrichment unit added. The methane enrichment unit is based on the presently developed Halder-Topsoe process. This unit contains only one methanator with recycle and the resulting gas has the heating value of 450 Btu/Scf. Schematic process Block Flow Diagram is presented in Figure 2 and 3, which show two alternate cases for producing 450 Btu/Scf gas.

Figure 4 shows the Grout Apfelbeck plot indicating the types of similar coals that have been economically gasified in commercial plants. In general, carbonaceous materials with an oxygen content above 10% show good reactivity for gasification. However, coals with rising carbon content could preferably be used for steam and power generation.

FIG 2:
SIMPLIFIED BLOCK FLOW DIAGRAM
METHANE ENRICHED FUEL GAS PLANT — ALTERNATE A

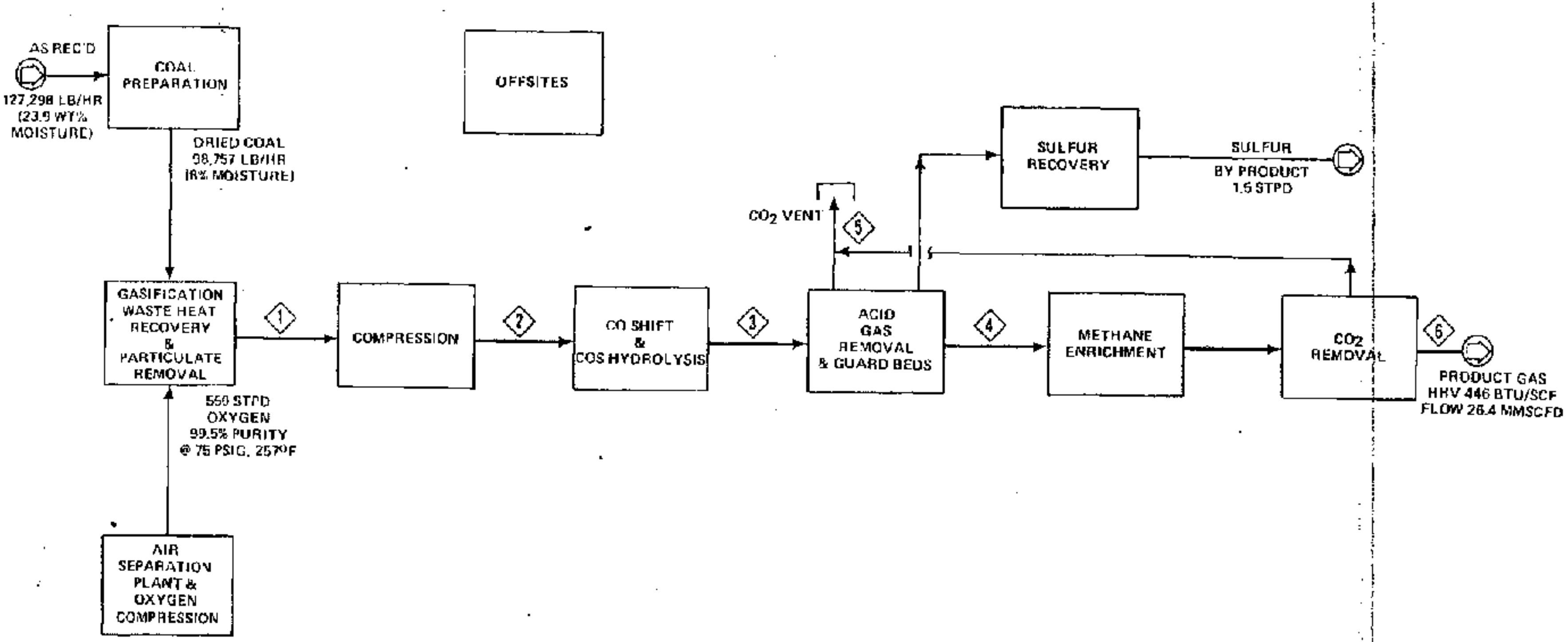
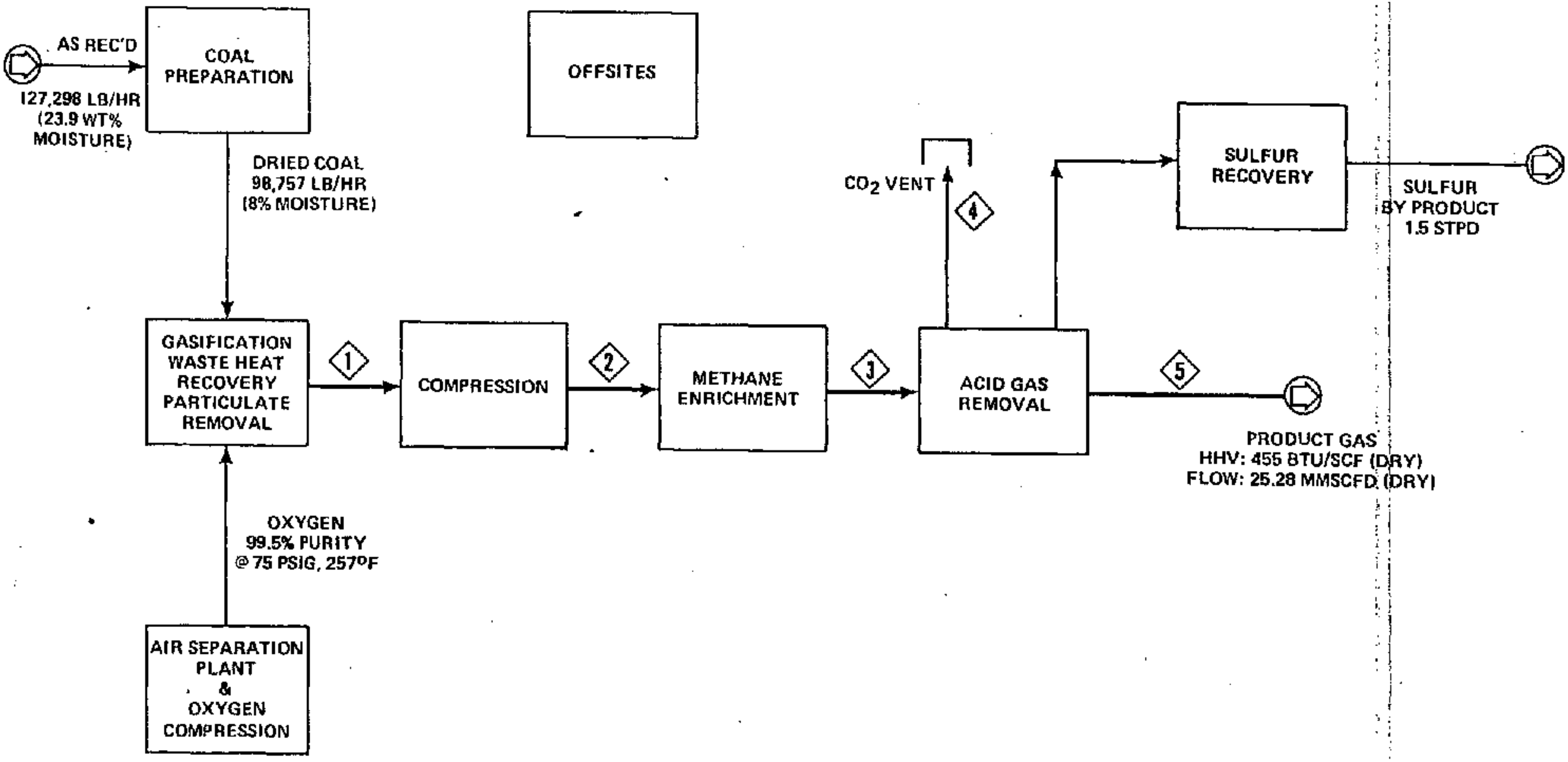
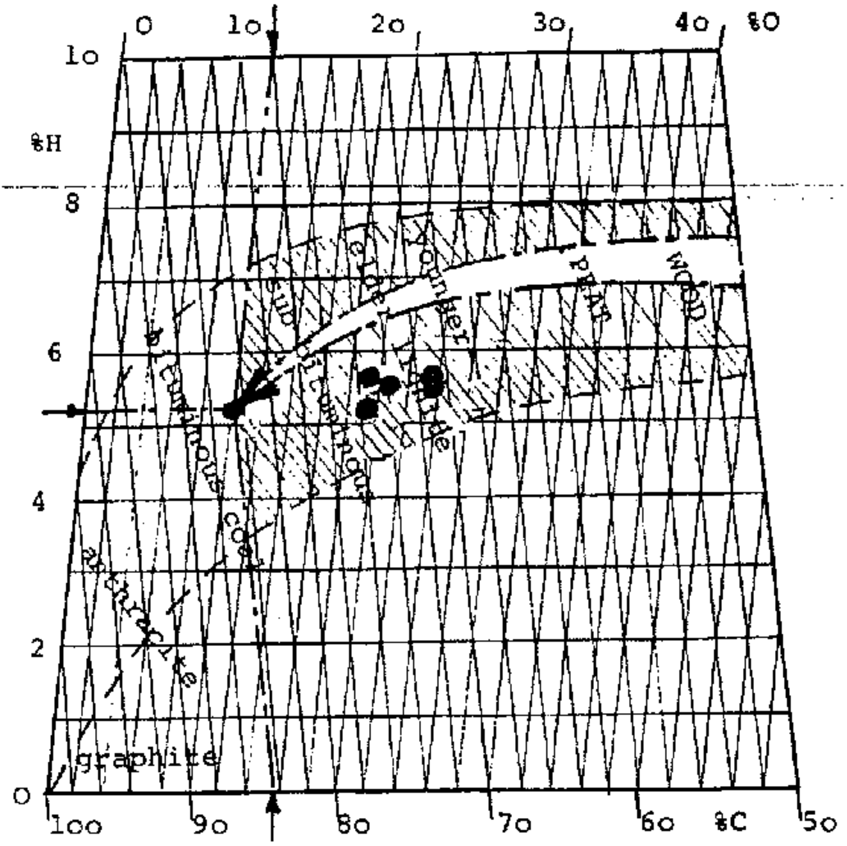


FIG 3:
SIMPLIFIED BLOCK FLOW DIAGRAM
METHANE ENRICHMENT – ALTERNATE B





STEAM - GASIFICATION COALS

Classification of carbonaceous material diagram by Grout-Apfelbeck ( C+H+O = 1oo %wt.)

← Economic feedstocks for Winkler fluid-bed gasification

| ○ Typical gasification coals | C | + | H | + | O | = | 100 % |
|---|---|---|---|---|---|---|---|
| Texas lignite | 71.4 | | 5.5 | | 23.1 | | |
| North Dakota brown coal | 71.5 | | 5.5 | | 23.o | | |
| Alaskan brown coal | 73.9 | | 5.5 | | 20.6 | | |
| Wyoming sub-bituminous coal | 75.5 | | 5.2 | | 19.3 | | |
| Brown coal of central Germany | 75.o | | 5.7 | | 19.3 | | |

-FIGURE 4-

The process described in this Section is a grass roots facility located near the coal supply. The coal considered is the Alaskan coal with the property as shown in Table 3.

Table 3 - Alaskan Coal Process Data

As Received Coal:

| Analysis | Wt.% | | |
|---|---|---|---|
| Carbon | 40.5 | HHV Btu/Lb (MAF): | 12409 |
| Hydrogen | 3.0 | Fusion Properties of Ash°F | |
| Oxygen | 11.2 | (Reducing Atmosphere) | |
| Nitrogen | 0.5 | Initial Deformation: | 2200 |
| Sulfur | 0.2 | Softening: | 2460 |
| Ash | 20.7 | Fluid: | 2535 |
| Moisture | 23.9 | F.S.I. = 0 | |

## Process Description

Coal Handling and Preparation Run-of-mine coal, which is 6" x 0, is received from unit trains unloaded, stacked and stored in 15 day capacity storage piles. The coal is retrieved from active storage and transferred to the process plant.

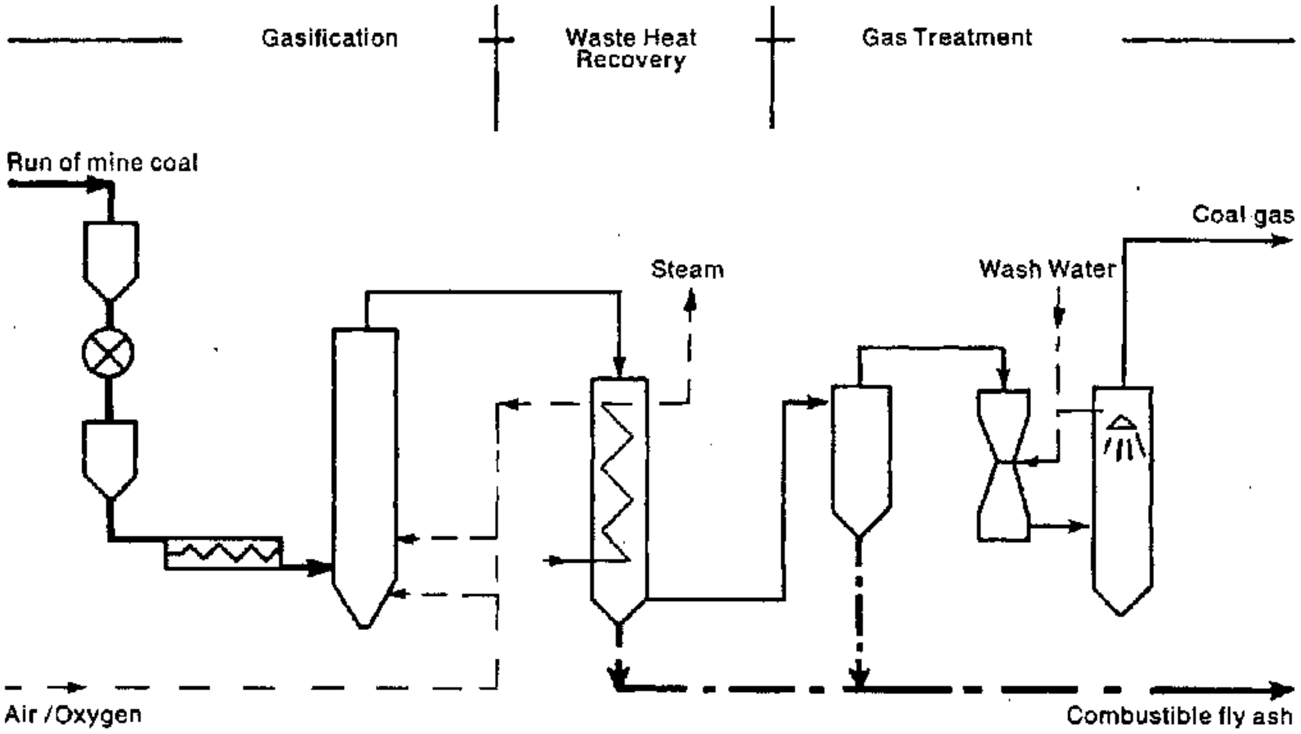
Coal to be used in gasification is crushed to 3/8" x 0, dried to 8% moisture and sent to active, closed storage ready to be transferred to the gasification section.

Gasification The gasification section consists of the gasifier, wasteheat recovery system and particulate removal system. The gasifier operates at four atmospheres and 2100°F. The coal is fed into the gasifier by means of a lockhopper system. The coal feed rate is controlled by variable speed screw conveyors. A simplified process Flow Diagram is shown in Figure 5.

The gasifier is a brick-lined, cylindrical shaft equipped with nozzles for the injection of air or oxygen and steam which acts as a medium for fluidizing the fine granular coal, subjecting the whole fuel bed to turbulent motion.

The air, or an oxygen/steam mixture, is also introduced into the space above the fluidized bed to ensure more complete gasification of coal particles entrained in the upward flow of gaseous products.

The Davy fluidized bed gasification process*



Winkler fluid-bed gasification unit
simplified process flow-diagram

- FIGURE 5 -

Depending on the ash softening temperature and reactivity of the fuel, the gasification temperature ranges in general between 1500-2200°F; at these temperatures reforming of volatile matter in the fuel is complete and no tar or liquid by-products are produced.

Larger ash particles fall to the bottom of the generator and are removed by a discharge screw but the greater part of the ash is carried off in the gas stream.

During gasification, the carbon in the coal reacts with oxygen and steam to form carbon oxides, methane and hydrogen. Oxygen is supplied by air separation plant. This plant also supplies nitrogen for process and utility use. The hot raw gas exists the top of the gasifier. Residue solids are removed from the bottom of the gasifier, and finer particles exit with the gas. The ash exiting from the bottom is lockhoppered to atmospheric pressure and pneumatically conveyed to storage for eventual disposal.

The hot gases leaving the gasifier enter the waste heat recovery system. This unit uses the heat in the gas to generate high pressure, superheated steam which is used throughout the plant. The solids, entrained with the raw gas, are a combustible char (approximately 30% carbon, 70% ash) which can be used as fuel in the utility boilers. A portion of the char is collected at the bottom of the waste heat recovery unit but the majority is removed in a dry cyclone following the waste heat recovery system. The dry char collected from the waste heat sections and cyclones are conveyed to the battery limits.

Fine particulates that may still be entrained in the gas are removed via a direct quench venturi-type scrubber. The particle laden water is sent to settling ponds where the particulate is concentrated, filtered, and the water sent to waste water treatment for water recovery.

Preparation of 450 Btu/Scf Gas The methane enrichment is based on operating around 250 psia. The raw particulate free gas is first compressed to 280 psia in centrifugal turbine driven compressors.

Two methane enrichment processes are considered in this paper. Process Alternate "A" involves the Standard Methanation Catalyst where the inlet gases for the reaction should have sulfides limited to 10 ppb. This process includes the standard upgrading of hydrogen in the raw Winkler gas using the sulfer tolerant CO shift catalyst, the $CO_2$ & $H_2S$ removal, followed by

ZnO guard beds for the final sulfides removal to 10 ppb level.

The Alternate "B" process deals with the direct methanation of the compressed raw Winkler gas using newly developed sulfide tolerant methanation catalyst by Haldor-Topsoe.

Alternate A - 450 Btu/Scf Gas Production The compressed Winkler gas enters the sulphur tolerant CO-shift unit where the ratio of Hydrogen to Carbon monoxide is adjusted to the required level for single stage methanator. Upon leaving the CO shift reactor, the gas passes through a COS hydrolysis unit. The raw gas contains a certain amount of carbonly sulfide (COS) which must be removed in order to prevent any emission problem in the subsequent acid gas removal unit. This is achieved by reacting the COS with steam to produce hydrogen sulfide which can be controlled more effectively in the acid gas removal unit.

From the COS hydrolysis unit, the gas enters the acid gas removal unit (Selexol) to remove the total sulfides to ppm level and adjust the $CO_2$ content in the gas. The gases from the $H_2S$ stripper are sent to the sulfur recovery unit (Stretford type) where sulfur is obtained as by-product. The gas leaving the $CO_2$ stripper, containing mainly $CO_2$, is simply vented to the atmosphere through stack.

The synthesis gas leaving the Selexol unit contains only trace levels of $H_2S$ and COS and has the proper ratio of CO, $CO_2$, and $H_2$ for methanation. Guard beds are provided for removal of trace sulfur compounds before methane-enrichment unit.

The methanation of the synthesis gas is an essential step in the manufacture of fuel gas with 450 Btu/Scf from high concentration of carbon oxides. The following reactions occur:

$$CO + 3\,H_2 \qquad CH_4 + H_2O$$

$$CO_2 + 4\,H_2 \qquad CH_4 + 2H_2O$$

The process is characterized by its simple and economic design and high energy recovery with recycle operation, done by means of an ejector[8]. The ejector system is a feature of the Halder-Topsoe Methanation Process (TREMP)[9].

The product gas from the methanation unit is cooled and sent to the second $CO_2$ removal unit in the Selexol system for final control of $CO_2$ content in the product gas.

Alternate B - 450 Btu/Scf Fuel Gas Production The main process steps involved in the production of the fuel gas is further simplified by having newly developed Halder-Topsoe sulphur tolerant methanation catalyst. A pilot plant operation of the sulphur tolerant methanation catalyst is now scheduled for autmn 1981 at Frederikssund, Denmark[10].

The compressed gas is sent directly to the methanation-enrichment unit containing the sulfur tolerant catalyst. This process permits methanation of the sulfur containing compressed raw gas from a Winkler gasifier and requires no additional steam. The contained water vapor in the compressed gas is quite enough for the reaction. The following reactions take place:

$$CO + H_2O \qquad CO_2 + H_2$$

$$2CO + 2H_2 \qquad CH_4 + CO_2$$

The gases from the methanation-enrichment unit are cooled and sent to the Selexol acid gas removal unit where $H_2S$ and $CO_2$ are removed to the required level.

## Comparison of the Methane Enrichment Alternatives

A summary of technical data and investment cost estimates are given in the following table for the two alternatives.

Table 4: Comparative Tabulation for Methane-Enrichment Medium Btu Gas Production

| Consumption Data | | Alternative A | Alternative B |
|---|---|---|---|
| Coal | mt/h | 58 | 58 |
| Power | Kwh/h | 13,800 | 12,650 |
| Cooling Water | m3/h | 2,380 | 2,220 |
| BFW | m3/h | 55 | 55 |
| M. P. Steam | t/h | 29 | 17.5 |
| Refrigeration requirements | $10^6$ Btu/h | 4 | 4 |
| **Product Rates** | | | |
| Product Gas | MMSCFD | 26 | 25.3 |
| | Billion Btu/Day | 11.7 | 11.7 |
| Char* | Billion Btu/Day | 2 | 2 |
| H. P. Steam | mt/h | 70 | 52 |
| Sulphur | kg/h | 1,400 | 1,400 |
| **Cost Estimate** | | | |
| Plant Investment | US $ | 40 million | 37 million |

*This char from Winkler is combustible and can be utilized in combination with low heating value gas or coal to generate additional H.P-steam.

Table 5 shows the material balance for a plant producing 11.7 billion Btu/day methane enriched fuel gas using the Alternatives A & B.

## Conclusion

This development of an enrichment methane gas provides an economically attractive alternative for the substitution of fuel oil for boilers, furnaces, and is also suitable for town and industrial gas.

## Table 5: Material Balance For Methane Enriched Fuel Gas Plant

ALTERNATE ROUTE - A

| Stream Number | 1 | | 2 | | 3 | | 4 | | 5 | | 6 | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| Stream Name | PARTICULATE FREE GAS | | COMPRESSED GAS | | GAS TO ACID GAS REMOVAL | | GAS TO METHANE ENRICHMENT | | $CO_2$ VENT | | PRODUCT GAS | |
| Component | MOL% | LBMOL/HR | MOL% | LBMOL/HR | MOL% | LBMOL/HR | MOL% | LBMOL/HR | MOL% | LBMOL/HR | MOL% | LBMOL/HR |
| CO | 39.36 | 2161.24 | 40.05 | 2055.84 | 10.63 | 688.58 | 15.17 | 687.50 | 0.09 | 1.83 | 9.80 | 286.00 |
| $CO_2$ | 18.33 | 1006.75 | 18.66 | 957.65 | 35.89 | 2324.96 | 9.10 | 412.43 | 99.74 | 2156.03 | 4.28 | 124.18 |
| $H_2$ | 35.18 | 1931.82 | 35.80 | 1837.61 | 49.48 | 3204.87 | 70.69 | 3203.79 | 0.07 | 1.62 | 62.64 | 1816.00 |
| $CH_4$ | 3.02 | 165.65 | 3.07 | 157.57 | 2.43 | 157.57 | 3.45 | 156.50 | 0.09 | 2.01 | 20.80 | 603.06 |
| $N_2$ | 1.38 | 75.84 | 1.41 | 72.17 | 1.12 | 72.17 | 1.59 | 72.00 | 80PPM | 0.17 | 2.48 | 72.00 |
| $H_2S$ | 0.07 | 3.74 | 0.07 | 3.56 | 0.06 | 4.06 | - | - | 9PPM | 0.02 | - | - |
| COS | 0.01 | 0.54 | 0.01 | 0.51 | 2PPM | 0.01 | - | - | 5PPM | 0.01 | - | - |
| $H_2O$ | 2.65 | 145.28 | 0.93 | 47.79 | 0.39 | 25.64 | - | - | - | - | - | - |
| Total | 100.00 | 5490.86 | 100.00 | 5132.70 | 100.00 | 6477.86 | 100.00 | 4532.22 | 100.00 | 2161.69 | 100.00 | 2899.24 |
| Press, psia | 40 | | 280 | | 240 | | 230 | | 20 | | 200 | |
| Temp, °F | 104 | | 335 | | 100 | | 100 | | 70 | | 70 | |
| HHV, BTU/SCF (DRY) | 279 | | 279 | | 220 | | 314 | | - | | 446 | |
| FLOW, SCFH (DRY) | 2,028,590 | | 1,929,672 | | 2,448,553 | | 1,719,932 | | 820,340 | | 1,100,231 | |

ALTERNATE ROUTE - B

| Stream Number | 1 | | 2 | | 3 | | 4 | | 5 | |
|---|---|---|---|---|---|---|---|---|---|---|
| Stream Name | PARTICULATE FREE GAS | | COMPRESSED GAS | | METHANE ENRICHED GAS TO ACID GAS REMOVAL | | $CO_2$ VENT | | PRODUCT GAS | |
| Component | MOL% | LBMOL/HR | MOL% | LBMOL/HR | MOL% | LBMOL/HR | MOL% | LBMOL/HR | MOL% | LBMOL/HR |
| CO | 39.36 | 2161.24 | 40.05 | 2055.84 | 22.60 | 911.15 | 0.09 | 1.15 | 32.79 | 910.00 |
| $CO_2$ | 18.33 | 1006.75 | 18.66 | 957.65 | 38.42 | 1549.61 | 99.72 | 1234.12 | 11.37 | 315.49 |
| $H_2$ | 35.18 | 1931.82 | 35.80 | 1837.61 | 19.08 | 769.38 | 0.11 | 1.38 | 27.67 | 768.00 |
| $CH_4$ | 3.02 | 165.65 | 3.07 | 157.57 | 17.63 | 710.80 | 0.06 | 0.80 | 25.58 | 710.00 |
| $N_2$ | 1.38 | 75.84 | 1.41 | 72.17 | 1.79 | 72.17 | 0.01 | 0.17 | 2.59 | 72.00 |
| $H_2S$ | 0.07 | 3.74 | 0.07 | 3.56 | 0.09 | 4.06 | 8PPM | 0.01 | - | - |
| COS | 0.01 | 0.54 | 0.01 | 0.51 | 0.01 | 0.01 | 8PPM | 0.01 | - | - |
| $H_2O$ | 2.65 | 145.28 | 0.93 | 47.79 | 0.38 | 15.33 | - | | - | - |
| Total | 100.00 | 5490.86 | 100.00 | 5132.70 | 100.00 | 4032.51 | 100.00 | 1237.64 | 100.00 | 2775.49 |
| Press, psia | 40 | | 280 | | 250 | | 20 | | 240 | |
| Temp, °F | 104 | | 335 | | 100 | | 70 | | 70 | |
| HHV, BTU/SCF (DRY) | 279 | | 279 | | 315 | | - | | 460 | |
| SCFH (DRY) | 2,028,590 | | 1,929,672 | | 1,524,480 | | 469,672 | | 1,053,270 | |

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