SECTION IV
CS/R HYDROGASIFICATION PROCESS

SECTION IV
ASSESSMENT OF CS/R HYDROGASIFICATION PROCESS

1.0 SUMMARY

The assessed process is a conceptual complex by Rockwell which integrates the Rockwell SRT Hydrogasifier with other more conventional units to produce an SNG product at a rate of 250 billion Btu/day and a co-product of benzene at 448 T/D.

Meetings and discussions with the developer were conducted to gather data on the status of development, test results, conceptual block flow diagram and material balances. From these several process strengths/weaknesses, potential improvements, required component development, critical questions and recommendations for further activity were derived.

The Rockwell material balance is keyed to a ratio of 0.2 lbs H_2/lb M.F. coal fed to the hydrogasifier, a carbon conversion of 5.3% to benzene, 45% to methane and 45% unconverted char. Although the balance around the hydrogasifier is based upon considerable tests results, the overall plant material balance is strongly influenced by factored estimates for other units without the benefit of a detailed design. This is especially true in areas such as utility plants and oxygasification which are wholly or partially fed by coal. As a result the overall cold gas thermal efficiency of 58% should be viewed as an early estimate, and this may increase significantly when the complex is optimized.

Critical areas in question as the overall process is now conceived are the H_2/Coal feed ratio, final process selected for H_2 production, the degree of co-product benzene production as it affects the final economics, addressing scale-up designs for commercial level, realistic expectations of operating factors and turn down.

The H_2 production process is yet to be selected ranging from candidates such as Texaco's partial oxidation process to an SRT dry fed char oxygasifier yet to be developed.

The strengths of the CS/R process as proposed appear to be high carbon conversion to CH_4 in the gasifier, accepting broad range of feeds, the option of a valuable co-product with no tars and high throughput. A large H_2 recycle system and oxygen plant might be considered as the principal weaknesses.

Some potential improvements suggested as a result of this assessment are: optimized benzene coproduct option; a more economical H_2 separation process; use of catalyst, and reduced H_2/Coal ratio.

Several components or elements which we envision to be important and required for the final commercial development are dense phase lockhopper feed systems, control/safety systems, hot solids flow measurement and control, hot solids-in-gas heat exchange, dry hot char separation and feeding and integrating gasifier units.

Following are further descriptions of the subjects summarized above.

2.0 CURRENT STATUS OF DEVELOPMENT

2.1 Program Background Chronology

Rockwell originally made a proposal to the Office of Coal Research (OCR) in 1974 which resulted in a coal liquefaction contract starting in 1975. In 1976 the dense phase coal feed system was demonstrated and a 1/4-TPH and a 1-TPH liquefaction reactor testing was started. In 1977 the coal gasification program was started along with a 1-TPH coal liquefaction PDU. In 1978 long duration gasification tests were started and a 4-TPH gasification program initiated. In 1979 the 4-TPH gasification program was redirected by DOE replacing the 4-TPH hydrogasifier reactor development facility with a 3/4-TPH integrated process development unit (IPDU).

2.2 Hydroliquefaction Program

This program was conducted under Contract No. EX-76-C-01-2044 (DOE) for $4,250,000. The period of performance was originally 49 months extending from August 1975 to September 1979 but was extended through February 1980. A three-year follow-on program is currently planned. The objectives are:

- Demonstrate Dense Phase Feeding
- Demonstrate Injector Mixing
- 1-TPH Engineering Scale Tests
- Construct/Operate a 1-TPH PDU to establish quantity and quality of liquid yields.

2.3 High Btu Hydrogasification Programs

The first contract, EX-77-C-01-2518 (DOE), for $1,500,000 from February 1977 to August 1978 (17 months) had the objectives of:

- Bench scale testing at Cities Service R & D Company
- 1/4-TPH engineering scale tests
- Preliminary process analysis

The second on-going contract, EX-78-C-01-3125 (DOE) for $22,000,000 from September 1978 to June 1982 (42 months) has for an overall objective the further development of the Rockwell single-stage short-residence-time hydrogasifier to demonstration plant status. Special objectives are to:

- Design, construct, and operate a 3/4-TPH coal feed rate integrated process development unit; demonstrate same in a 30-day test (continuous operation).

- Develop process data and operating experience to support design, economic evaluation, and optimization of a viable commercial process.

- Prepare a preliminary design of a practical commercial plant.

The program scope of work involves an integrated combination of design, construction, and operation to demonstrate the feasibility of the Rockwell hydrogasifier reactor for commercial application.

Testing is currently being performed at 3/4-TPH in a short- duration engineering-scale facility to improve and refine the process data base by generating essential information outside the scope of the previous contract.

Process conditions are being directed towards optimum benzene production as a co-product with SNG.

It was initially planned that a 4-TPH unit be developed and used to make extended runs for various durations up to 30 days. However, in August 1979, DOE decided to redirect the program, replacing the 4-TPH hydrogasifier reactor development facility with a 3/4-TPH integrated process development unit (IPDU). Lengthy tests will be performed to demonstrate system operability, component durability and product quality, while shorter runs will be conducted to evaluate process factors. It will no longer be possible to investigate injection element scaling, as was originally planned, by studying single-element and clustered multiple-element injectors.

Tests will be made with strongly caking bituminous coal as well as with subbituminous coal; char from each will be characterized. Problems with the process, materials and operation shall be defined and resolved to the extent necessary to warrant low-risk go-ahead with a demonstration plant venture following completion of this project. A preliminary design of a commercial-scale plant of such quality and detail as to be directly useful to an Architect/Engineer firm in the final design of an actual commercial unit is the ultimate output of the project.

2.4 Data Base

The data base resulting from the above work covers three facilities and a broad parametric range. The three facilities and resulting data points were:

- Cities Service Bench Scale - 58 data points
- Rockwell 1/4-TPH Hydrogasification - 49 data points
- Rockwell 1-TPH Hydroliquefaction -110 data points

The parametric ranges were:

- Bituminous, subbituminous coal and peat
- Residence times 30-5000 msec
- Pressures 500-1500 psi

- Temperatures	1400-2000 F
- H_2/Coal ratios	0.25-1.0
- Coal throughput	1000-3000 lb/hr

Rockwell has judged the data from the different facilities to be consistent at the full range of coal throughput (References 1 & 2).

2.5 Commercial Hydrogasification Reactor Operating Conditions

The range of hydrogasifier operating conditions which are applicable in the design of a commercial SNG plant using either bituminous or subbituminous coals, or peat, include:

- Residence Times	1000-3000 msec
- Pressures	500-1500 psia
- Temperatures	1700-1900 F
- H_2/Coal Ratios	0.20-0.45
- Commercial Reactor Throughput	140-220 ton/hr.

3.0 PROCESS DESCRIPTION

The following describes the developer's Preliminary Commercial Concept Design of the Cities Service/Rockwell (CS/R) Coal Hydrogasification Process to produce SNG from coal at a rate of 250 billion Btu/day (HHV) and benzene, a principal liquid co- product. (Refer to Process Block Flow Diagram, Fig. IV-1).

3.1 Summary

The key features of this process are: an entrained flow short-residence-time (SRT) coal hydrogasifier reacting coal with hot hydrogen to a raw gas high in methane content and to char; an entrained flow, char/coal/oxygen/steam gasifier for the production of hydrogen make-up; and a cryogenic hydrogen-methane separation system yielding an SNG product and recycle H_2.

Because of the high initial carbon conversion to methane in the hydrogasifier only a trim methanation subsequent step is required.

By controlling the temperature and the residence time of the reactants in the coal hydrogasifier, benzene is also produced as a valuable coproduct.

Both the coal and char gasifiers are fed using a dense-phase dry solids system using H_2 or other reacting gas as the transport medium.

The balance of the process units in the plant are conventional consisting of: coal preparation, oxygen plant, gas quench, benzene recovery, shift conversion, acid gas removal, trim methanation, sour water stripping, ammonia recovery, sulfur recovery, solids/liquids effluent recovery, steam/power/water treatment and off sites.

Feed materials consist of coal, raw water and air. Products are High-Btu Gas (SNG) and Benzene. By-products are sulfur and ammonia. Effluents are CO_2, clean flue gas, and solids sludge (mainly ash) and water losses.

3.2 Coal Preparation/Feeding

The raw coal is prepared conventionally by crushing to 70% minus 200 mesh and dried to about 2% moisture.

The prepared coal is fed to the gasifiers in dense phase using H_2 or other transport gas through a pressurized, two-stage, cycling lock hopper. The coal to the steam power boilers is fed by standard dilute phase pneumatic means.

The coal analysis in the proposed Rockwell process using Pittsburgh Seam No. 8 is as follows:

	Wt.% Dry
C	71.50
H	5.02
N	1.23
S	4.42
Ash	11.30
O (by difference)	6.53

3.3 Oxygen Plant

The oxygen plant is comprised of commercially available air separation units from which liquid O_2 is pumped to the reactor at a pressure of approximately 1000 psig. The O_2 is vaporized by heating to either 200F for the hydrograsifier or 300F for the char oxygasifier.

3.4 Coal Hydrogasification

This unit consists of three reactor trains, or modules, each employing an entrained flow reactor utilizing a rocket-engine-type injector scheme. Rocket engine injector design techniques are used to achieve rapid and thorough mixing of the pulverized coal and hot H_2. Each module is comprised of a preburner, injector, reactor and heat recuperator, all of which are integrated within a pressurized shell.

The preburner's function is to raise the recycle + make up gas (about 92 vol. % H_2) to the required temperature from 1500F by reacting with O_2. The gas then passes to the injector.

In the injector, coal (2% moisture, 70% - 200 mesh)in dense phase is fed using hydrogen as transport gas (approx. 0.003 lb. mols per lb. of coal). The coal rapidly mixes with the heated H_2 gas from the preburner at the reactor inlet to achieve a theoretical mixed temperature of 1400F. The hydropyrolysis reactions are carried out in the reactor section. The product gases are subsequently cooled in the recuperator section directly beneath.

In the reactor the coal is both pyrolyzed and reacted with the hydrogen gas. The products are dependent upon the residence time, initial temperature, pressure and reactant composition and can vary from a totally gaseous product to one in which significant amounts of high purity benzene are co-produced. In the case described the variables are set to produce a "moderate" level of benzene (nominally 5.8% carbon conversion to benzene) as well as about 26% CH_4 in the raw gas. The H_2 recycle rate is set at 0.20 lbs H_2/lb MF coal leaving an excess of H_2 carried with the raw product gas. Most of the excess H_2 is recovered in the cryogenic separation unit and recycled.

The overall reaction can be broadly expressed as follows:

$$\text{Coal} + H_2 = CH_4 + C_6H_6 + C_2H_6 + CO + CO_2 + H_2 + NH_3 + H_2O + \text{Char}$$

The reactor conditions based upon Rockwell 1/2-TPH bituminous coal test No. 313-23 are:

H_2 preburner inlet temp	1500F
Coal/O_2 inlet temp	200F
O_2 inlet temp	200F
Reactor outlet temp	1772F
Recuperator exit temp	834F
Reactor pressure	1000 psig
Residence time	2470 msec

The overall carbon conversion based on regression analysis of all Rockwell 1/4-TPH hydrogasifier bituminous coal tests and feeding Eastern Bituminous Pittsburgh Seam No. 8 coal is taken as 55.0%. The carbon distribution is as follows:

5.8% to C_6H_6, 45.4% to CH_4, 3.25% to CO, 0.42% to CO_2, 0.14% to C_2H_6, and 45.0% residual in char.

The char stream is then separated from the raw product gas via several stages of cyclones and is subsequently fed to the char/coal oxygasifier unit.

3.5 Gas Treatment and H_2 Recovery

The raw product gas which has been separated from the char and quenched is then processed through the following steps:

- o Benzene Solvent Absorption
- o Diglycol Amine Acid Gas Removal
- o Trim Methanation
- o Gas Drying
- o Cryogenic Methane/H_2 Separation

The SNG product gas exiting the cryogenic unit is compressed to 1000 psig and 120F and has a composition of approx. 94.5% CH_4 and 4.8% H_2, with the balance consisting of N_2, AR, ethane, CO and only traces (0.3 ppm) of H_2S.

3.6 Hydrogen (Make-up) Production

Hydrogen is produced by reacting the char from the hydrogasifier with steam and oxygen to produce a raw syngas mixture of H_2, CO, CO_2, H_2S, some methane and ammonia. The char is supplemented with coal to produce the required H_2 quantity to balance the plant.

The char coal gasifier is a pressurized, entrained flow, short-residence-time oxygasification reactor. The same dense phase solid transport feeding techniques used in the hydrogasifier will be employed in the oxygasifier to maximize reactor thermal efficiency. Several candidate configurations for this application include those under development by Texaco, Shell-Koppers and Mountain Fuel Resources.

The overall reaction is broadly expressed as:

$$\text{Char} + \text{Coal} + O_2 + H_2O = CO + H_2 + CO_2 + H_2S + CH_4 \\ + NH_3 + N_2 + H_2O\ \text{(excess)}$$

Reactor conditions are targeted at:

Reactor pressure = 950 psig
Reactor exit temp = 2460F
Char inlet temp = 834F
Steam inlet temp = 1000F
O_2 inlet temp = 300F
Coal inlet temp = 200F

Performance data were determined from kinetic and equilibrium calculations at the stated exit reactor conditions.

The raw syngas is further processed by conventional steps as follows:

- Gas Quench & Solids Removal
- Shift Conversion
- Acid Gas Removal
- Trim Methanation

The resulting make up gas consists of about 87.8% H_2, 4.6% CH_4 and 7.6% H_2O.

3.7 Other Operations

The other operations consist of conventional effluent treatment and byproduct recovery steps such as solids (ash) recovery, sour water stripping, ammonia recovery, sulfur recovery and bio- oxidation.

The utilities plant includes water treatment, steam and power generation and other facilities. Since this complex is conceived to be self-supporting from a utilities standpoint, the only major imports are raw water and coal.

3.8 General

The overall process described above is conceptual at this point. The material balance is based upon Rockwell experimental data around the hydrogasifier to a large extent and around the oxygasifier. In addition, the product gas treatment from the acid gas removal through cryogenic methane separation is based upon a screening type evaluation study performed by Air Products and Chemicals, Inc., and reported in May 1979.(3)

The balance of the plant units do not have the benefit of an engineering design from which a detailed closed material balance could be generated. As a result much of the treatment units and the utility sections have been factored into this preliminary balance. Because of this it is suggested that comparative conclusions not be drawn for such items as overall plant thermal efficiency et.al., since power generation and heat recovery play significant roles in such factors. However, the process thermal efficiency may be viewed with much greater confidence since this can be derived from the principal process stream material balance which is based on broad experimental background.

4.0 STRENGTHS AND WEAKNESSES

The items listed and briefly described below are considered to be strong points and weak points characteristic of the CS/R Hydrogasification Process relative to current commercial or near commercial processes for producing high Btu SNG.

4.1 Strengths

- <u>High Carbon to Methane Conversion</u>

  A relatively high single pass carbon conversion (45%) to methane in the gasifier reduces the downstream conversion requirements using only trim methanation.

- <u>Versatile Fuel Application</u>

  This type of gasifier has been found to be applicable to all types of coal and other solid fuels such as peat and lignite.

- <u>No Catalyst Required</u>

  No catalyst addition or recovery systems required.

- <u>Dense Phase Feed</u>

  Dry fed dense phase coal using reactant transport gas reduces heat requirement relatively to a slurry feed.

- <u>High Btu Gas with Liquid Option</u>

  The end product may be adjusted from all gas to benzene coproduction providing an attractive potential flexibility.

- <u>High Throughput Rates (Short-Residence-Time) and Small Reactor Size</u>

  Relative to reactor volume this type of reactor allows around 2000 lb/hr/ft^3 of coal feed. This is reflected in the short residence time (seconds or less) and the small reactor size.

  The small reactor size results in many advantages which might be unavailable or impractical for larger conventional reactors.

The highest quality of materials of construction may be used since they are not such an economic factor.

Use of modular reactor elements allows ease of transition from pilot to commercial scale including testing at full element or cluster size.

Most of the key reactor components can be shop fabricated and tested under high quality assurance levels. Maintenance and replacement of elements is simplified and downtime theoretically reduced.

Small reactant inventory allows rapid shutdown or quench.

- <u>No Tars in Raw Product Gas</u>
  The absence of liquid hydrocarbons and tars allows almost complete vapor phase product recovery processing steps. The benzene is recovered by liquid absorption and purification.

4.2 Weaknesses

- <u>Large H_2 & Recycle System</u>
  An excess of H_2 is needed to satisfy the CH_4 synthesis and coal heat up to reaction temperature. This H_2 is carried through the raw SNG processing steps, cryogenic separation and recycle.

- <u>O_2 Plant Required</u>
  The production of H_2 for make up requires a large O_2 plant.

- <u>Lockhopper System</u>
  Coal and char fed by dense phase which is an advantage still requires a high pressure (1000 psi) lockhopper system.

- <u>Key Steps Require Development</u>
  Several areas require further development to validate the overall process concept as follows:

- Char oxygasifier
- Heat recovery of solids + gas streams
- Scale up from modular to full scale reactors
- Feed splitting to modular elements
- SRT control/safety systems

5.0 POTENTIAL IMPROVEMENTS

The following items are suggested as potential solutions to problem areas that appear to exist as the processes are now proposed by the developers. These also take into account the stage of development of the overall process and the conceptual status of many of the companion operations to and around the proposed gasification step. These suggestions are the result of reviewing items mentioned in other sections such as weaknesses, alternates, disadvantages and status of development. It is suggested that these are potentials only viewed from the perspective of this assessment and will require more detailed investigation and evaluation prior to testing. It is for this reason they are called potential improvements.

Some potential improvements which appeared worthy of investigation are as follows:

1. Increase BTX (Benzene) production to a maximum.
2. Reduce the H_2/coal (or carbon) ratio to a practical minimum.
3. Apply catalysts to the gasification step(s).
4. Apply a H_2 separation process (such as Monsanto hollow fiber process) to recover H_2 for recycle to reduce processes downstream of gasification and acid gas removal.
5. Process all the coal through the hydrogasifier producing a larger quantity of char. Use char only for H_2 production and any

balance for utility steam/power generation. A larger portion of the total coal volatiles would be captured in the raw product gas and less lost to CO_2 in the steam boilers and to some extent in the char gasifier.

6. Burn a stream of desulfurized raw product gas or a stream of raw syngas from the char gasifier for steam/power generation to reduce need for flue gas desulfurization.

7. Produce hydrogen by reforming a portion of the product methane rather than oxygasification of char and coal.

5.1 Increase BTX (Benzene) Production to a Maximum

The base case considers benzene production at a moderate level of 5.34% based on carbon converted in the hydrogasifier. Under conditions to produce a maximum of this coproduct the yield can double.

On the basis of value placed on purified recovered benzene by Rockwell, this increase would further credit operating costs by another $62.2 million annually. This would reduce the gas cost by $0.76 per million Btu.

5.2 Reduction of the H_2 to Coal or Carbon Ratio to a Minimum

The CS/R coal gasification process as proposed by the developers for this assessment has fixed the H_2/coal ratio in the hydrogasifier feed at 0.2041 lbs H_2/lb MF coal. Experimental runs were made at ratios from 0.25 to 1.0. The stoichiometric quantity of H_2 converted (gasifier + methanation) is 0.0904 lbs of H_2/lb MF coal with 0.128 lbs H_2 exiting with the raw product gas to be recovered and recycled. The optimum (minimum) level of H_2 to coal has not been established but is being approached gradually by ongoing testing.

For purposes of assessing the magnitude of cost reduction due to a decrease in the H_2/coal ratio, it is assumed that the same conversion could be achieved at a H_2/coal ratio midway from the stoichiometric requirement and the level proposed. This would be 0.147 lbs H_2/coal and the following reductions could be expected:

(a) Reduced gas flow through quench
(b) Reduced gas flow through benzene recovery
(c) Reduced gas flow through acid gas removal
(d) Reduced gas flow through methanation
(e) Reduced gas flow through drying and cryogenics
(f) Reduced recovered hydrogen flow
(g) Reduced total recycle flow.

Rough estimates of the percentage capital cost reduction and the cost effect on gas cost for the above are as follows:

	% Reduction	Total Capital Cost Reduction ($$10^6$)
(a)	17.	9.7
(b)	17.	2.3
(c)	17.	3.1
(d)	25	11.3
(e)	25	17.8
(f)	25	
(g)	12	
		44.2

Effect on Gas Cost = $0.10/$10^6$ Btu reduction

5.3 Apply Catalyst to the Gasification Steps

Work in areas other than SRT (short residence time) coal gasifiers using low cost catalyst additives to enhance the conversion and reactivity of coal suggests that there may be potential to apply catalysts to the Rockwell CS/R Hydrogasification Process as well. Although experimental data are lacking for SRT applications certain advantages drawn from other processes appear to merit investigation.

Some advantages envisioned are:

- Possible reduction of H_2/C ratio with proportionate cost reductions to all operations related to H_2/C level.
- Lower gasification temperatures favoring higher CH_4 yields at a given H_2/C ratio in the hydrogasifier.
- Reduced downstream process steps after gasification depending on the reduction of recycle and excess components in the raw product gases.

Some of the added steps which would have to be more than compensated by the benefits would be:

- Catalyst cost and consumption
- Catalyst addition operations
- Catalyst recovery
- Spent catalyst disposal

Since a measure of this potential is not available for an SRT gasifier at this time, a quantitative estimate of the benefits has not been attempted and this discussion is limited to qualitative items above. However, it is judged that if catalyst addition proves to be justified the overall reductions should be at least equal to that of minimizing the H_2/C ratio described above.

5.4 Apply an Improved H_2 Separation Process

A significant portion of the energy consumed in the Rockwell CS/R Hydrogasification Process is related to the separation, recovery and recycling of a large stream of H_2 from the raw product gas. The proposed process employs a cryogenic separation and recovery system.

The potential exists for cost reductions if a lower energy consuming and simpler operation could be devised and applied to this process. One candidate process might be the Monsanto Prism® hollow fiber H_2 separation process.

It appears that the pressure levels, H_2 partial pressure and other conditions of the raw product gas after acid gas removal present an ideal application for trial of the Monsanto process. The process has been applied successfully by the developer on a commercial scale for almost three years to processes under similar conditions.

The most obvious advantages appear to be:

- Elimination or significant reduction of the cryogenic operation.
- Reduced gas flow volume downstream of acid gas removal with corresponding reductions to methanation and gas drying units.

Rough calculations applying Monsanto published data and estimated recoveries of H_2 indicate that by applying this process, after acid gas removal, to the raw product gas the H_2 can be separated for recycle, the cryogenic section can be eliminated, the gas volume through methanation and gas drying is reduced to about half and the residual pressure remaining in the product gas approximately compensates for the recompression required for the recycle H_2. A more rigorous analysis is required before recommending testing for this application.

An estimate of the cost effect shows about equal total capital required. Any gain would be achieved by reduced operating costs mainly in energy savings around the cryogenic unit and gas compression. This would be reflected in coal consumption for steam/power generation. No estimate was attempted since the Rockwell conceptual process did not include a utility/power breakdown by process unit.

5.5 Process Total Coal Through Hydrogasifier

Rather than three separate coal feeds to the hydrogasifier, the oxygasifier and the steam/power generator, it was thought that there may be an advantage in processing all of the coal first through the hydrogasifier and using the resulting char to feed the oxygasifier and steam/power generator.

Some potential advantages envisioned were:

- o The capture of a larger proportion of volatiles which are lost to CO_2 as the process is now proposed where about 33% of the total coal to the plant goes to the oxygasifier and steam/power generator.
- o Recovering a higher level of BTX (benzene) in proportion to the SNG product since all of the coal volatiles will exit the hydrogasifier for recovery.
- o Little or no sulfur in the boiler flue gas. Using char fuel the sulfur would be reduced to 25% of coal fuel. Using a stream of desulfurized raw product gas the sulfur would have already been removed as H_2S in the acid gas removal unit.

It becomes obvious that hydrogasifying sufficient coal under the same conditions and conversion parameters to produce enough char for both H_2 production and boiler feed would lead to an excess of SNG. Possibly the way a balanced plant would be achieved would be to hydrogasify that quantity of coal to produce char for all H_2 required and using a slipstream of raw product gas after desulfurization as boiler fuel to balance the plant.

In addition, since a greater rate of coal is seen by the hydrogasifier for a given amount of SNG the H_2 quantity must be about the same as the quantity used in the process assessed in this study.

Using a thermodynamic equilibrium calculation program for the hydrogasifier developed in Lewis Research Center by McBride and Gordon(4), the calculations showed a net increase of about 13.0% total coal consumption reflecting an increased gas cost of about $0.23 per million Btu. The increase in benzene production was only 8% with an off- setting effect on gas cost of only $0.06/million Btu.

5.6 Burn Raw Product Gas for Power/Steam Generation

This alternate is aimed at eliminating the need for the costly flue gas desulfurization step required if the feed coal at 4.42% sulfur were burned as in the base case.

In order to achieve this a stream of raw product gas would be tapped after acid gas removal. To replace this gas quantity additional coal would be processed through the hydrogasifier.

This alternate is actually similar in part to Alternate No. 5 where the total coal feed is processed through the hydrogasifier including that required to supplement the char to the oxygasifier. Taken alone and still feeding coal to the oxygasifer about half of the effects might be realized. Since Alternate No. 5 proved to be a cost increase this alternate would also be more costly and was abandoned.

5.7 Two schemes were considered to produce hydrogen by reforming part of the product methane rather than oxygasification of char and coal. One scheme used the char/coal as fuel to the methane reformer. A second scheme used additional methane as fuel to the reformer.

The second scheme was rejected on the basis of signficantly higher capital costs. The hydrogasification stream and the hydrogen plant stream called for about twice the capital costs. Also, although the operating costs were not evaluated, there would be an excess of char which would add to operating costs and resulting overall product gas cost.

The first scheme was estimated to be about equal in capital cost to the base case. It was also rejected on the basis of probable higher operating costs since the hydrogasification plant was about double the size of the base case requiring a proportionate increase in coal feed. In addition, it required the use of a char/coal fired reformer which has not been developed.

6.0 COMPONENTS REQUIRING DEVELOPMENT

In order to assess the CS/R Hydrogasification Process from the perspective of a fully developed commercial scale operation several new operations auxiliary to the principal gasification steps must also be considered. These may not be apparent during the pilot scale development but when expanded to the required commercial scale the need for development becomes more obvious.

Some of these auxiliary operations which are critical to the successful performance of the conceptual design as proposed include:

(1) A full scale dense phase coal or char feed system.

(2) A fail-safe control system for hydrogen preheating and feeding to hydrogasification.

(3) A char separation, handling and dense phase feed system for oxygasification with steam.

(4) A hot char feeder control system under high pressure into an oxygen atmosphere reactor.

6.1 Dense Phase Coal Feed System (Commercial Scale)

A rough calculation indicates that pulverized coal at a relatively low bulk density requires a significant number of large high pressure vessels and a sizable recompression and letdown conservation system for transport gas to accommodate the proposed dense phase feed system.

Assuming an hourly cycling of the hopper feed vessels feeding into a 1000 psi reactor, limiting vessel sizes to 12 feet diameter, using recycle H_2 gas for transport medium and allowing some excess pressure residual in the evacuated feed vessels, approximately 25 vessels would be required designed for over 1200 psi in a high hydrogen partial pressure vapor phase.

Although the transport gas is used in the reactor and figures in the overall material balance, about twice this quantity will remain behind when the feed vessel has exhausted its coal. This gas must be removed, purged, recompressed and transferred to another vessel filled with coal to repeat the cycle.

The total bulk volume of coal amounts to about 25,000 cubic feet per hour. The void fraction containing the high pressure gas amounts to more than half of this volume. Translated to standard cubic feet per minute of circulating H_2 gas this is about 20,000 SCFM.

The possible impacts, response intervals, mechanical failures and hazards of such a system must be thoroughly investigated to insure that the reliability and safety will be in accordance with the 90% operating factor set for the commercial facility.

6.2 A Reliable Safe Gasifier Feed and H_2 Preheating System

An SRT high mass throughput gasifier system introduces requirements for advanced techniques of control safety and reliability when applied to a commercial scale not yet available from existing technology. To a partial extent rocket feed and control technology is certainly appropriate where applicable. In a coal hydrogasifier however the products discharge to a very large delicately balanced closed system with enormous inertia.

Precedents already exist in coal oxygasifiers being constructed on a large scale with failures in operation due to unrecognized inadequacies in unproven feed mechanism designs. These gasifiers were not short residence time high mass throughput which tends to compound the potential control problems. The transfer of components from conventional systems to new applications should be viewed with caution since their incompatibility may easily escape discovery.

While SRT reactors offer many real potential advantages they also introduce other potential problems. The most readily recognized seem to center on the area of control, reliability, response time. Some of these problems surface when viewed on a commercial scale by anticipating possible upset conditions and the requirements for safe and orderly recovery. Some of these relative to the gasifier are:

Upstream Upsets

- Loss of Coal Feed - unreacted H_2 to system, temperature drop
- Loss of H_2 Feed - unreacted O_2 to system
- Coal Feed Splitting to Modules

Downstream Upsets

- Char Plugging - Immediate over pressure and shutdown
- Loss of Quench - Over temperature and shutdown
- Pressure Buildup
- Relief System Failure

Reactor Upsets

- O_2/H_2 Balance - O_2 to system or temperature drop
- Module Balancing
- Average Conditions vs. Individual Stream Sensing
- Sound Attenuation and Mechanical Sonic Effects

Looking at this limited list of upsets the most obvious of the requirements seems to center around the problem of the very short time which will exist to sense, measure and effectively react before a failure or unwanted condition prevails. The following are some of the requirements which must be satisfied after first determining what and how fast an upset condition may occur:

- Anticipatory Sensing
- Direct Measurement Sensing
- Response Rate
- Recovery Rate
- Isolation
- Relief
- Safe Orderly Shutdown
- Provide Surge Capacity

The time intervals and inventories within and around the gasifiers are so small that the sensing and control function will probably not allow supervisory confirmation or response. This means that almost total control loop functions must be integrated within an instrumentation system with very high reliability and possibly high redundancy.

6.3 Hot Char Handling Separation & Dense Phase Feed to Gasifier

One of the possible alternates to the proposed process is the dense phase feeding of hot dry char to the oxygasifier for the production of the required hydrogen. Such a system does not exist and has not been tested in combination with the hydrogasifier.

The integration of such a system will require the development of the following components to operate continuously with the rest of the process:

- An efficient hot char separation and intermediate surge capacity.
- Dense phase feed system possibly with the introduction of supplemental transport gas if the raw product gas carried with the char is not sufficient or at high enough pressure. This will require an isolation method to operate the dense phase char feeding in a cycling lockhopper system similar to the coal feed to the hydrogasifier.
- A method of combining and balancing of a supplemental coal feed with the char if this is required as the proposed process indicates. If the coal and the char oxygasifiers are separate trains the product gases must be compatible as to the overall process so either train may be shutdown without shutting down the whole process, or a large over-capacity from each train may be required to carry the load temporarily.
- Possibly develop the oxygasifier to operate on either coal or char or a combination of coal plus char. This will require a variable steam-oxygen feed for optimum oxygasification and may introduce other feed, balancing and control problems.

6.4 Dense Phase Hot Char, Steam, Oxygen Feed Control System

The dense phase feeding of hot char poses the same design problems as noted for the dense phase coal feed system on a commercial scale but the following present additional problems which must be confronted:

- A safe system of control in all modes, i.e., start-up, shutdown, emergency and normal transient operations. This is particularly important when a large proportion of the reactant is high pressure oxygen.

- The possiblity of catastrophic explosion with high concentration of O_2 available to the oxygasifier due to several upset causes (loss of steam, loss of char feed, etc.).

- A reliable solids flow measurement and control system with high sensitivity, response and recovery.

- An injector mechanism functioning similar to the hydrogasifier injector but which will accommodate both O_2 and steam safely and reliably coupled with its control system.

7.0 REFERENCES

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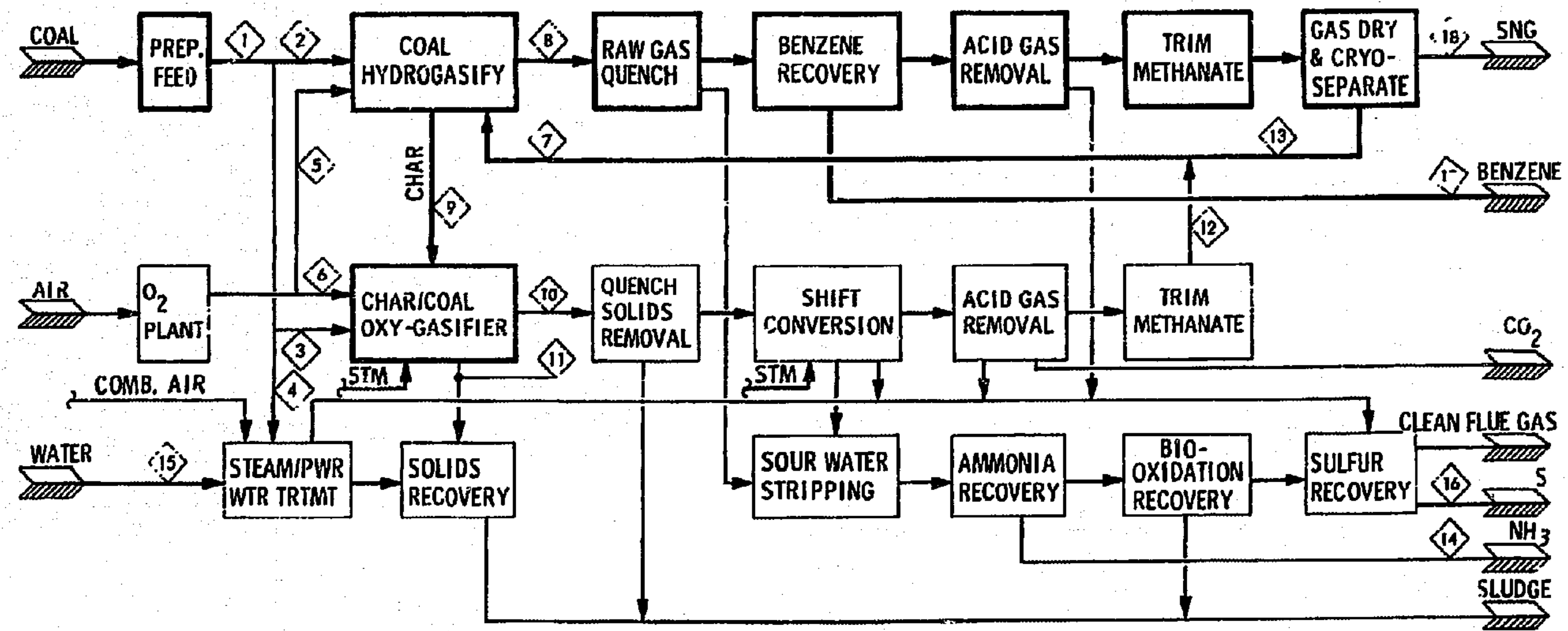


FEED		
COAL (2% MOIST)	16,528	T/D
O_2 (100%)	6,034	T/D
RAW WATER	67,596	T/D

PRODUCT		
SNG	$\approx 250 \times 10^9$	Btu/D
BENZENE	448	T/D
NH_3	123	T/D
S	738	LT/D

FIGURE IV-1. CS/R Hydrogasification Block Flow Diagram.

TABLE IV-1. CS/R Hydrogasification Process Material Flow Balance

STREAM - NO. ◇	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18
COMPONENT / - NAME	TOTAL COAL (2% MOIST)	HYDRO-GAS COAL	OXY-GAS COAL	UTILITY COAL	HYDRO-GAS O_2	OXY-GAS O_2	RECYCLE HYDRO-GAS	RAW GAS	CHAR	RAW SYNGAS	ASH	HYDRO-GAS MAKE-UP	RECOVERED HYDRO GAS	NH_3	RAW WATER	SULFUR	BENZENE	SNG PRODUCT
LB MOL/HR																		
CH_4							4,743	28,971		15		1,389	3,354					26,855
C_2H_6								38										38
C_6H_6								478									478	
H_2							93,497	57,436		19,211		42,929	50,567					655
CO								1,738		29,745								
CO_2								225		6,302								
H_2S								1,127		668								
NH_3								631		1				602				
N_2							202	202		128		128	73					128
AR					7	71	88	95		72		71	17					79
O_2					1,459	14,253												
H_2O	1,528	1,020	240	268			1,381	6,627		12,513		1,381						
TOTAL LB MOL/HR					1,466	14,324	99,911	97,568		68,656		45,899	54,012	602			478	27,755
TOTAL LB/HR	27,535	18,388	4,327	4,820	46,980	458,947	299,001	855,791		1,404,301		140,326	158,675	10,252	5,633,000		37,313	439,943
SOLIDS, LB/HR																		
COAL (MF)	1,349,799	901,383	212,116	236,300														
CHAR									405,622									
ASH											126,220							
SULFUR																68,920		
TOTAL OVERALL, LB/HR	1,377,334	919,771	216,443	241,120	46,980	458,947	299,001	855,791		1,404,301		140,326	158,676	10,252	5,633,000	68,920	37,313	439,943
TEMP, °F		200	200		200	300	70-1500	834	834	2,460	2,460		95					105
PRESS., PSIA		>1,000	>965		>1,000	>965	>1,000	<1,000	<1,000	<965	< 965	>400	388					1,000

SECTION V

ASSESSMENT OF THE EXXON CCG PROCESS

1.0 SUMMARY

The Exxon catalytic coal gasification process consists essentially of a single step gasifier which generates methane. The reactions in the gasifier involve coal gasification, steam-shift, and methanation. Therefore, there is no need for additional steps outside the gasifier for shift and methanation to produce SNG.

The coal feed is impregnated with K_2CO_3 catalyst prior to entering the gasifier. Approximately 90 percent of the carbon in the coal is converted to methane and carbon-dioxide in the gasifier; 10 percent of the carbon is converted to char and remains with the ash and catalyst. About 90 percent of the catalyst is recovered for reuse in the process. The residue ash/char/catalyst mixture is then disposed of.

From the gasifier raw gas SNG is separated from CO and H_2 which are then recycled to the gasifier. In the gasifier, recycled CO and H_2 are methanated while more CO and H_2 are produced from coal gasification. When the process operates at a steady state, the rate of CO/H_2 recycling equals the rate of CO/H_2 production resulting in a net production of CH_4 and CO_2 in the gasifier.

2.0 CURRENT STATUS OF DEVELOPMENT

Exxon Research and Engineering Company is engaged in research and development on a catalytic coal gasification (CCG) process for the production of substitute natural gas (SNG) from coal. An outline of the stages of development is shown below:

2.1 Previous Research Work

Sponsor:	Exxon
Period:	Pre-1979
Objectives:	General CCG concept and catalyst recovery using bench-scale units.

2.2 Predevelopment Program

Sponsor: DOE, $2.4 MM, Contract No. E(49-18)-2369

Period: July 1, 1976 through December 31, 1977.

Objective:

- o Operation of 6" x 30' fluidized bed gasifier (FBG) with Illinois coal.
  - Operate with mixed K_2CO_3/ Na_2CO_3 catalyst
  - Operate with recycled catalyst

- o Bench-scale studies on gasification kinetics and catalyst recovery.
  - Broaden data base to other coals
  - Test reactivity of recovered catalyst
  - Study critical factors in catalyst recovery
  - Operate the small fluidized bed Continuous Gasification Unit (CGU) and fixed-bed units to obtain additional kinetic data.

- o Conceptual design of a commercial CCG plant.
  - Continue engineering screening studies
  - Prepare an updated commercial plant study design.

During the Predevelopment Program several technical questions were resolved, and the technology has now moved into the Process Development Program.

2.3 Process Development Program

Sponsor: DOE/GRI, $16.8 MM
Exxon, $3 MM

Period: July 1, 1978 through June 30, 1981

Objectives: o Operation of 1-T/D Process Development Unit (PDU).

  - start up of gasification, gas separation, and catalyst recovery systems
  - Operate PDU as a total plant integrating every process system in the unit

- Bench research and pilot plant support.
- Engineering research and development.
  - Study economics and guide research
  - Define key features of the process for supporting engineering design and operations of a Large Pilot Plant (LPP).

Exxon disclosed in March, 1980, that the PDU construction was almost completed. Individual units had been tested for preparation of an integral PDU operation. During the test, it was found that when the gasifier was operated at 500 psi level, the bulk density of the fluidized bed was about 1/3 of the density which would be expected at the 100 psi pressure level.

After considerable development efforts, Exxon was able to adjust the expected density in the fluidized bed (e.g., by varying methods of catalyst impregnation and drying, etc.) and operate the gasifier at about 300 psig.

In May 1980 Exxon announced plans to construct a 100 TPD LPP in Holland with operation expected to begin by mid-1985.

Further details and discussion of the development program are contained at the end of this section.

3.0 Process Description

The following describes the process flow of the overall conceptual process plant of the Exxon Catalytic Coal Gasification Process by section as indicated in the block flow diagram, Figure V-1. The heat and material balance are taken from Exxon's Commercial Plant Study Design(1).

3.1 Coal Preparation and Catalyst Addition

The feed coal is crushed to minus 8 mesh size in the coal handling and storage section.

The feed coal as received by the plant is an Illinois No. 6 bituminous coal, washed, or cleaned, in a beneficiation plant at the mine. The ultimate analysis is as follows:

	Wt.% (dry)
C	69.67
H	5.05
O	9.45
N	1.84
S	4.19
Cl	0.08
Ash	9.72
	100.00

HHV (Btu/lb dry coal)	12,730
Moisture, Wt. % (as received)	16.5%

The feed coal from the storage is dried from 16.5 wt.% to 4 wt.% moisture in the entrained dryer using flue gas generated in a coal-fired burner as the drying medium. The dryer overhead stream, which contains hot vapor entrained with the dried coal is separated in the cyclone separator. The separated hot gas is recycled to the coal-fired burner except that a slip stream is vented to the flue gas desulfurization unit through an electrostatic precipitator for removal of solid fines. The dried coal separated from the cyclone is transferred via a screw conveyer to a zig-zag blender where the catalyst solution is added and mixed with the coal. The K_2CO_3 catalyst-soaked coal is then transferred to another entrained dryer where coal-fired burner flue gas is again employed as the drying medium to dry the catalyst impregnated coal. The overhead stream which contains the dried coal impregnated with catalyst and hot gas is routed to a cyclone separator. The separated hot gas from the cyclone is recycled to the coal-fired burner except that a slip stream is vented to the flue gas desulfurization unit after its solid fines are removed by an electrostatic precipitator. The separated coal from the cyclone is then transferred to a storage bin facility ready for feeding the gasifier.

3.2 Gasifier System

The catalyst-impregnated coal is transferred by gravity to the lockhopper feeding system which consists of the low pressure hopper, the lockhopper and the high pressure feeder. The recycle syngas containing carbon monoxide and hydrogen is employed as the pressure medium for the lockhopper system. After the coal is transferred from the low pressure hopper to the high pressure feeder, it is pneumatically carried into the gasifier in dense-phase flow by the preheated recycle syngas. The preheater is provided for superheating the steam-containing recycle syngas in the radiant section and for preheating a slip stream of the dry recycle syngas in the convection section. The latter is used as the carrier gas for coal feeding.

In the gasifier the catalyst-impregnated coal is fluidized by the superheated stream of the steam-containing recycle syngas. The steam reacts with the fluidized catalyzed coal char, in presence of the recycled syngas containing carbon monoxide and hydrogen. Methane and carbon dioxide as well as hydrogen sulfide and ammonia are produced.

The main reactions taking place in the gasifier are the highly endothermic steam gasification reaction, the mildly exothermic steam-gas shift reaction, and the highly exothermic methanation reaction. The steam-gas shift and methanation reactions are essentially at equilibrium over the catalyzed char in the gasifier. The composite of the three reactions results in no significant net production of carbon monoxide and hydrogen. The resulting overall reaction can be represented as follows:

$$\text{Coal} + H_2O = CH_4 + CO_2 + H_2S + NH_3$$

The above reaction is essentially thermo-neutral. Therefore, only a small amount of heat input to the gasifier is required, primarily to preheat the feed coal and to provide for heat losses. This heat requirement is supplied by the preheater via the coal feeding syngas stream and by the superheated steam-containing recycle syngas stream. The reaction temperature and pressure in the gasifier are maintained at 1275° and 500 psia, respectively.

The char and ash are withdrawn from the gasifier bottom to the char quench drum and then to the char slurry drum for catalyst recovery.

3.3 Heat Recovery and Gas Scrubbing

The product gas from the gasifier is first routed through two cyclone separators connected in series and integrated with the gasifier, and then through a series of heat exchangers for recovery of high temperature heat by generating and superheating high pressure (600 psia) steam required for the gasifier. After recovery of the high level heat, the process gas stream is still well above its water dew point. The bulk of solid fines contained in the process gas stream is then separated in the tertiary cyclone. The gas then proceeds to the process gas saturator and to the Venturi scrubber where the final clean up of solid fines is effected. The clean process gas is then routed through a series of heat exchangers for further heat recovery by preheating boiler feed water and generating low pressure (65 psig) steam. At some point of heat recovery when the process gas is cooled to about 330°F, it is passed through a fixed-bed reactor to catalytically hydrolyze carbonyl sulfide (COS) to H_2S. The remaining heat in the process gas stream is then rejected by the coolers. At the end of heat recovery, the process gas enters the ammonia scrubber at 120°F, wherein ammonia is removed from the gas.

3.4 Acid Gas Removal and Sulfur Recovery

The overhead gas stream from the ammonia scrubber is routed to the Selexol acid gas removal unit which primarily consists of the H_2S absorber, the H_2S stripper, the CO_2 absorber and the CO_2 stripper. The effluent stream from the H_2S stripper is routed to the sulfur recovery unit and the overhead stream from the CO_2 stripper is vented to the atmosphere. Low pressure steam is employed for reboiling the H_2S stripper. Air is used for stripping off CO_2 from the Selexol solution in the CO_2 stripper.

The sulfur recovery unit consists of the Claus plant and the tail gas treating plant. Sellable elemental sulfur is recovered in this unit as a byproduct.

3.5 SNG Separation

The treated process gas from the acid gas removal unit is then passed through the Molecular Sieve unit for drying and trace CO_2 removal. This preparation is required for feeding the downstream cryogenic SNG separation unit.

The process gas stream, now containing only methane, hydrogen and carbon monoxide is heat exchanged with various cold product streams from the cryogenic fractionation system, and is chilled down to a cryogenic temperature. A flash separation is provided at a cryogenic temperature to vaporize the major portion of CO_2 and H_2 from the liquid CH_4. The liquid from this flash separation is then fed to the cryogenic fractionation column operating at approximately 40 psi, where the final separation of methane from the remaining syngas is performed. The overhead gas stream containing primarily the syngas is heat exchanged with the feed stream and then routed to combine with the flashed syngas stream at the feed. The total combined syngas stream is then recycled through the preheater to the gasifier and the lockhopper system. The bottom product from the cryogenic fractionation column is also heat exchanged with the feed stream, vaporized and then compressed to the gas pipeline pressure for sales.

3.6 Sour Water Stripping and Ammonia Recovery

This section primarily consists of the sour slurry stripper, the H_2S/NH_3 stripper and the ammonia recovery system.

The sour slurry stream containing approximately 10% of solid fines is routed from the Venturi scrubbing system to the sour slurry stripper. The overhead vapor stream from the sour slurry stripper is fed to the sulfur recovery unit and the bottoms is routed to the filter belt press. The filter cake is transferred to disposal and the filtrate routed to the catalyst recovery system.

The sour condensate streams from various K.O. drums in the heat recovery and gas scrubbing section and the NH_3 scrubber bottoms are routed to the sour water stripper. The H_2S rich stream is separated by distillation from the NH_3 stream and fed to the sulfur recovery. Ammonia is recovered as 20 weight percent aqueous solution as a byproduct. The stripped water is routed to waste water treating.

3.7 Catalyst Recovery

This section consists of the $Ca(OH)_2$ digester and fourteen water-leaching stages. All the catalyst containing streams throughout the plant are first routed to the $Ca(OH)_2$ digester and then to the water-leaching stages for recovery of the catalyst. These streams include:

- The char from the bottom of the gasifier, after being slurried in the char drum,
- The solid fines from the tertiary cyclone separator, after being slurried in the fines slurry drum, and
- The filtrate of the sour slurry stripper bottoms.

Lime (CaO) and the makeup catalyst as 30 wt.% KOH are added to the $Ca(OH)_2$ digester for recovery of catalyst tied up with the coal minerals. Fresh catalyst makeup is necessary since a portion of the catalyst is not recoverable from the coal minerals.

The CaO is hydrolyzed in the digester to form $Ca(OH)_2$. The ratio of calcium in the lime feed, to potassium in the feed char and fines solid is 0.7 lb Ca/lb K. The char and fines slurries are soaked in the digester for two hours by agitation at 70 psia and 300°. Under these conditions, approximately 90% of the potassium in the feed solids is solubilized. The remaining potassium leaves with solids in water-insoluble compounds.

About 98.5% of the potassium salts solubilized in the $Ca(OH)_2$ digestion are recovered in the downstream water-leaching stages. Overall, this section recovers 87% of the total potassium catalyst which entered the gasifier with the feed coal. The remaining 13% is supplied by the makeup KOH.

All the recovered plus the fresh makeup catalyst, containing 37 wt.% K_2CO_3 equivalent, are then recycled to the catalyst addition section for impregnation of the feed coal.

3.8 The Plant Arrangement

The conceptual commercial process plant with a capacity of 250 MMSCFD product SNG is envisaged to consist of the following trains:

- Four trains of Coal Drying and Catalyst Addition (three trains normally in operation, one train spare);
- One train of Coal Storage Bins;
- Four trains of Reactors, except two trains of pressurization gas handling and one common spare train of feed hopper system and the lockhopper recycle gas compressor;
- Four trains of Acid Gas Removal and Sulfur Recovery;
- Two trains of SNG Separation system;
- Two trains of Sour Water Stripping and Ammonia Recovery;
- Two trains of Catalyst Recovery System.

4.0 STRENGTHS AND WEAKNESSES

The following are the strengths and the weaknesses of the Exxon Catalytic Coal Gasification Process.

4.1 Strengths

- <u>Simple Process Sequence</u>

  The gasification step combines the reactions of coal gasification, steam shift and methanation in one single gasifier vessel. The net products of the gasifier are methane and carbon dioxide. It therefore eliminates the requirement of having additional downstream processing steps of shift reaction and methanation for producing SNG.

- <u>High Carbon Conversion</u>

  The carbon conversion in the gasifier is estimated to be in the 90-percent range, and the primary products are methane and carbon dioxide.

- <u>Heat Integration/No Oxygen Plant</u>

  As the reaction in the gasifier is a combination of coal gasification, steam shift and methanation, the composite heat balances of these reactions are essentially neutral. The net chemistry of these reactions can be represented as follows:

  $$\text{Coal} + H_2O = CH_4 + CO_2 \qquad \Delta H \approx 0$$

  As indicated above, this reaction is almost thermally neutral and in fact only a small amount of heat is required in the gasifier to preheat the feed coal and to provide for heat losses. Due to this specific feature, an oxygen plant is not required, and potential slagging problems associated with oxygen use are eliminated.

- <u>No Caking Problem</u>

  No pretreatment is required for caking coals. The action of the catalyst to the pore structure of coal minimizes the caking problem associated with metaplast formation in the beginning of the coal-gas reactions.

- <u>Easy to Control</u>

  The gasification reaction in the gasifier reaches equilibrium. The residence time is not critical to the product compositions; therefore, the process is easy to control.

- **Simple Gasifier**

  The gasifier is a fluidized bed. No complicated special internals are required. Even temperature distribution in the gasifier is expected due to the fluidization mixing effects of the bed.

- **No Slagging/No Tar**

  As the gasifier reaction temperature is 1275°F, the slagging problem is eliminated. Due to the gasification temperature, exotic materials are not required for the gasifier construction.

- **Energy Efficient for CH_4 Recovery**

  The cryogenic system for separation of CH_4 from syngas uses pressure reduction to achieve auto-refrigeration required in the system. Additional heat exchange between the product streams and the feed stream to chill the feed stream to cryogenic temperature is provided. External mechanical refrigeration is not required for normal operation of the system.

- **Recovered Catalyst Activity Maintained**

  The experiments have indicated that the recovered catalyst maintains its catalytic effect over long recycle operation, as there were no indications of activity reduction.

- **Data Base**

  - During the Predevelopment Program, the fluid bed gasifier has demonstrated the following characteristics:

    a. Good quality data
    b. High on-stream factor
    c. Recycle catalyst as active as fresh catalyst
    d. Fluid-bed operation stable and easy to control
    e. High carbon and steam conversions with a simulated syngas recycle.

- A computerized mathematical model was developed for simulation of the gasifier operation. The simulated results agreed well with the observed data obtained from the 100 psig gasifier operation tests.

- During the Process Development Program, the Process Demonstration Unit (PDU) gasifier sustained 165 hours of stable operation with Illinois No. 6 coal feed in August, 1979 with the following conditions:

  a. 90 percent carbon conversion.
  b. Gasification at 1250°F and 500 psig.
  c. The bed fluidized with steam and N_2 gas.
  d. Fines returned to bed by cyclone.
  e. The run terminated by failure of liquid nitrogen pump.

The PDU work is currently in progress. The major effort of this work is to demonstrate a continuous stable operation for a period of time of the PDU (1 T/D capacity) which consists of all the process systems to be provided in a commercial plant. Also, an investigation will be made of the effect of trace components build-up in each system of the overall process operation.

4.2 Weaknesses (Areas Needing Development)

o Catalyst Recovery

The catalyst recovery system currently being contemplated will recover approximately 90 percent of the catalyst originally impregnated with the coal. The long residence time needed in the water-leaching catalyst recovery process indicates high investment on equipment. The catalyst tends to tie-up with aluminum compounds in coal forming water-insoluble salts. Therefore, if the feed coal contains a large amount of aluminum compounds, high rates of catalyst makeup will result.

o Gasifier

As the reactions in the gasifier are to reach equilibrium, a high residence time is required resulting in a big reactor vessel. Exxon is investigating ways for improvement in this area.

- **Digestion**
  $Ca(OH)_2$ digestion is required to recover the water-insoluble portion of the catalyst. However, the digestion step produces a lot of solid fines which contribute difficulties in solid/liquid separations in the downstream water-leaching process.

- **High Steam Requirement**
  The gasification needs a steam rate of 1.585 pounds of steam per pound of dry coal. In order to supplement the requirement, the off-site reboiler needs approximately 16 percent of the total plant coal feed as fuel for generating the required process steam.

- **10% Char Loss**
  The gasifier converts approximately 90 percent of carbon in the feed coal to gases. The remaining unconverted char is transferred with ashes to the catalyst recovery system and eventually disposed off-plot.

5.0 POTENTIAL IMPROVEMENTS

The following potential improvements for the Exxon CCG process are suggested from the perspective of this assessment and will require more detailed evaluation prior to testing. Some potentials which appear worthy of investigation are:

1. Improved Flue Gas Desulfurization
2. Improved Low Level Heat Recovery
3. Improved Cryogenic SNG Recovery
4. Improved Catalyst Recovery

5.1 Improved Flue Gas Desulfurization

Exxon's original process design showed two coal dryers: one for crushed feed coal, the other for catalyst impregnated coal. Both dryers use the hot flue gases generated from the coal-feed burners as the drying media. Since the flue gases are from the coal burners, they contain sulfur compounds; therefore, the flue gases from both dryers are routed to a desulfurization unit for making H_2SO_4 as a by-product.

A potential improvement might be to replace the coal generated flue gases by the flue gas from the recycle gas preheater. The preheater is fueled by the clean product SNG; therefore, its flue gas is environmentally clean. If this clean flue gas is used as the drying medium in each dryer, then the flue gas from the dryer can be vented to the atmosphere without being desulfurized, and the flue gas desulfurization unit can be eliminated.

As to the flue gas from the coal-burning offsite boiler, it might be routed to the tail-gas treating unit of the Claus sulfur recovery plant. A section of the treating unit could incorporate a hydrolyzer and an absorber for such treatment.

A preliminary cost estimate indicates that the additional equipment would cost about 30% more than that saved, but the coal feed saved by deletion of the coal burners would pay out in about four years. In addition, the separate H_2SO_4 storage, handling and shipping facilities could be deleted by elimination of the flue gas desulfurization unit.

5.2 Improved Low Level Heat Recovery

Exxon's original process design showed that the low level heat below 313°F in the process gas stream is rejected to both air and water coolers. The process gas is cooled to 120°F prior to entering an ammonia scrubbing column.

The proposed improvement is to utilize the low level heat currently rejected to air for generating refrigeration[2, 3]. This is made possible by providing an ammonia-absorption refrigeration unit. This unit would have a single stage absorber and the process gas stream would be used for providing desorption heat from 313°F to 288°F.

The refrigeration load would be used primarily in the acid gas removal unit (Selexol Process), and the dry-bed unit. The latter is provided for dehydrating the process gas stream prior to entering the cryogenic SNG separation unit.

As a part of the low level heat is recovered for generating refrigeration load, part of the heat rejected is reduced, resulting in savings on cooling surfaces as well.

A preliminary cost estimate indicates that the net capital cost for equipment added versus deleted is zero. However, there would be a utility savings by reduced power requirements of packaged refrigeration units and on air coolers.

5.3 Improved Cryogenic SNG Recovery

Exxon's original design was improved by an Air Products scheme. The improvements include elimination of the cascade refrigeration unit, and replacing a fractionation column with a stripping column. These improvements were made possible by pressure letdown of the methane containing gas. The effect of auto-refrigeration plus heat exchange was sufficient to condense the bulk of the methane. Flash separation and stripping at a lower pressure than the original design effect the separation of SNG (methane) from CO and H_2 gas.

Further improvements over the Air Product's scheme are suggested by JPL. These improvements include replacing the letdown valve upstream of the feed drum (to the stripper) with a turbo-expander. The latter will not only extract horsepower for driving the recycle and gas compressor, but also cool the process gas stream further. This will result in savings in operating horsepower. The second improvement suggested by JPL is to use a multi-stage LNG pump to deliver the pressure required for SNG product; namely, pump the LNG up to the 1000 psi level and vaporize the remaining liquid downstream at the battery limit. This will save SNG compressors and horsepower as well.

A preliminary cost estimate indicates that the equipment cost savings would be about $2,000,000 and utility power savings would be about $4,000,000 per year.

5.4 Improved Catalyst Recovery

The catalyst is known to associate with minerals in the coal to form water-insoluble compounds. The improvement concept is to remove some of the ash and minerals of the coal upstream of catalyst impregnation by beneficiation. Should this beneficiation prove feasible, the catalyst tie-up with the coal mineral would be reduced with the potential of increased recovery of the catalyst.

6.0 DEVELOPMENT STATUS DETAILS

6.1 Key Results From Previous Research Work

Previous Exxon sponsored research on catalytic coal gasification was performed in bench-scale units which have the capability of operating at pressures up to 1000 psig as well as in a small pilot-scale Fluid Bed Gasifier (FBG) unit with a coal feed capacity of up to 25 lbs/hr and a maximum operating pressure of 100 psig. This pressure limitation is present because the FBG was originally built for thermal gasification work. During 1975, the FBG Pilot Plant was operated with K_2CO_3 catalyzed Illinois coal for continuous periods of up to two weeks. Good quality data were obtained for yield periods covering a wide range of operating conditions. For many yield periods, the FBG operated with synthetic gas makeup (simulated recycle) such that inlet and outlet synthesis gas rates were in approximate balance.

Close approaches to gas methanation equilibrium were demonstrated with K_2CO_3 catalyst in both bench-scale units and the FBG pilot plant. Bench-scale rate data were obtained for Illinois coal with both K_2CO_3 and Na_2CO_3/K_2CO_3 catalysts. These data were combined with analytical descriptions of fluid bed contacting to develop a first-pass computer model of a fluid bed catalytic gasification reactor.

In the area of catalyst recovery, the effectiveness of a water wash for recovering about two-thirds of the catalyst was demonstrated. The forms of this recovered catalyst were identified and work was initiated on the recovery of water- insoluble catalyst. Also during this phase, engineering screening studies were carried out for commercial plants to establish preferred configurations for process flow and equipment sequencing, and to determine investment and operating costs.

6.2 Key Results from Predevelopment Program

6.2.1 Fluid Bed Gasifier (FBG) Operation

The continuous operation of the 6" x 31' fluid bed gasifier (FBG) was to simulate all commercial gasifier parameters except pressure, the effect of recycle gas rate, and the resulting effect on reaction kinetics. These parameters which were representative of expected commercial conditions include type of coal, coal size distribution, catalyst loading, reaction temperature, steam conversion, carbon conversion, fluidizing velocity, residue composition, bed density, and fluidization properties of the gasified solids. Results from the FBG operations are summarized below:

(a) The unit was used to develop fifty material balanced periods. Of these, eighteen were selected to represent a variety of process variables for detailed workup. Unit operations were of high quality. The service factor during the last six months of operation averaged more than 80% of real time, with a one-month maximum of 96%.

(b) FBG Operations confirmed the ineffectiveness of mixed sodium and potassium catalyst.

(c) Operations using recycled water soluble catalyst reached a recovery level of 94% of water soluble potassium (64% of total potassium). After approximately ten cycles of operation with recovered catalyst, no loss of activity nor any significant buildup of other constituents was observed. Pilot scale calcium digestion experiments demonstrated recovery of more than 90% of the total potassium from FBG residue. Recycle of catalyst at this recovery level will be a part of the development phase.

6.2.2 Bench-Scale Studies

The bench-scale research activities generated several significant results as follows:

(a) A computer model of the rate controlling reaction kinetics was developed. It describes gasification rate as a function of temperature, catalyst loading, and gas composition. Pressure is important only because it influences gas composition.

(b) Early testing of mixed sodium-potassium catalyst indicated that this system would be ineffective for reducing catalyst cost.

(c) The effort was redirected toward increasing the recovery level of the water-insoluble potassium. The most promising approach is the reaction of ash/char residue with calcium hydroxide to produce soluble potassium salts and insoluble calcium aluminum silicates. This reaction is carried out in an aqueous digestion system at relatively mild conditions. It results in an increase in catalyst recovery from about 70% with no calcium hydroxide to about 90% with ratios of calcium to potassium of the order of 0.7 moles/mole.

(d) Exposure of char to air was found to oxidize sulfides to sulfates and to inhibit the effectiveness of the water wash. Calcium digestion in the presence of CO was observed to convert some of the potassium sulfate to potassium formate.

(e) Potassium sulfide was found to be catalytically active but less effective than the hydroxide and carbonate forms when the gasifying medium is pure steam. The carbonate and hydroxide forms are equal in effectiveness.

(f) Wyoming subbituminous coal was found to be kinetically equivalent to Illinois bituminous coal in the presence of potassium catalyst.

6.2.3 Engineering Screening Studies

The engineering screening studies led to the following major conclusions:

(a) The preferred form of makeup catalyst for catalytic gasification is potassium hydroxide (KOH) manufactured by electrolysis of potassium chloride (KCl). Reserves of KCl in North America are very large relative to the amounts needed. Because KOH for catalytic gasification would be produced in relatively large quantities and low purities over a long term, the cost could be significantly below the current market price.

(b) With KOH at the current market price, calcium hydroxide digestion to recover water insoluble catalyst from spent gasifier solids is justified in addition to water washing to recover water soluble catalyst.

(c) The addition of a secondary gasification step to raise carbon conversion to 95% from the base level of 90% provides only a marginal economical incentive.

(d) The selective Selexol scrubbing process for acid gas removal is somewhat lower in cost than scrubbing with non-selective hot potassium carbonate or selective refrigerated methanol.

6.2.4 Commercial Plant Study Design

The engineering research and development efforts culminated with the preparation of a new Catalytic Coal Gasification Commercial Plant Study Design. The process bases for the Study Design were set based on the results of the laboratory and engineering studies carried out during the Predevelopment Program. The key findings of the Study Design are:

(a) The estimated total investment for a pioneer commercial plant feeding Illinois No. 6 coal and producing 257 billion Btu per stream day of substitute natural gas (SNG) is $1,530 million. This is for a January, 1978 cost level at an Eastern Illinois location. A "process development allowance" and a "project contingency" are included in this estimate, consistent with standard Exxon practices.

(b) The estimated cost of SNG produced from this pioneer gasification plant is \$6.20 per million Btu (\$/MMBtu). This gas cost is an initial selling price based on 100% equity financing, a 15% DCF return, and escalation rates of 6% per year for SNG revenues and 5% per year for net operating costs. On an alternative financing basis of 70% debt/30% equity with 9% interest on debt, the comparable initial gas cost is \$4.70 per MMBtu.

(c) Several factors could reduce the SNG cost below the Study Design range of \$4.70-6.20/MMBtu. These include larger plant capacities, surface-mined coals, increased government financial incentives, and future savings based on the learning experience gained from the pioneer plant and from further research and development.

The Study Design economics are believed to be a realistic prediction of the costs (in 1978 dollars) for a pioneer commercial plant. Caution must be used when comparing these economics with published estimates for other coal gasification processes. Such estimates can vary widely depending on the process, offsites, and economic bases, the investment estimate approach, and the maturity of the technology. It is expected that a consistent comparison with state-of-the-art gasification technology will show a significant incentive for further development of the Catalytic Coal Gasification Process.

The details on the Predevelopment Program of the CCG process have been documented in the Final Project Report on Predevelopment Program for Exxon Catalytic Coal Gasification Process dated December, 1978 prepared by Exxon Research and Engineering Company, Baytown, Texas(1).

6.3 Process Development Program

This program is currently in progress. Work plans and the current status are summarized in the following sections.

6.3.1 Overall Milestone Schedule

The overall project milestone schedule for the SNG Program is included as Figure V-2. It shows the following four major tasks from 1978 through 1981.

a. PDU Startup
b. Integrated PDU Operation
c. Bench Research and Pilot Plant Support
d. Engineering Research and Development

6.3.2 Status of Process Development Unit (PDU)

As of October, 1979, the PDU status is reported as follows:

(a) Sustained 165 hours of stable operation with Illinois No. 6 coal feed and steam in unit from August 10 to 17, 1979.

- 80 to 90% carbon conversion
- 1250°F and 500 psig
- Fluidized with steam and nitrogen
- Fines returned to bed by cyclone
- Run terminated by loss of liquid N_2 pump.

(b) Manually removed char as a water slurry.
- Activated automatic system.

(c) H_2 and CO supply systems ready for use in next run on a once-through basis.

(d) Gas cleanup and recycle system nearly ready.
- MEA and molecular sieve systems complete and in startup operation.

- Cryogenic fractionator in final assembly.

(e) Catalyst recovery construction underway.

6.3.3. CCG Development Issues

The following summarizes various CCG issues to be resolved in different stages of the development.

Issues	Bench	PDU	LPP
o Fluid Bed Gasification			
- Lockhopper Feed System	X	X	X
- Gasification Reaction Rate	X	X	-
- Fluid Solid Contacting	-	X	X
- Properties of Steady-state Char	X	X	-
- Fines Generation	-	X	X
o Gas Recycle Loop			
- Preheat Furnace	X	X	-
- Cryogenic Separation	X	-	-
- Trace Components Build-up	-	X	-
o Catalyst Addition and Recycle Loop			
- Trace Components Build-up	X	X	-
- Fines Generation	X	X	-
- Solid-Liquid Separation	X	X	X
- Solid Disposal	X	X	X
- Water Wash vs. $Ca(OH)_2$ Digestion	X	X	-

6.3 4 Research Studies Planned for 1979

The following items are the research efforts planned for 1979.

(a) Catalyst Loop Research

- Costs of concentrating dilute catalyst solutions by evaporation.
- Evaluation of alternative solid-liquid separation methods.
- Evaluation of tradeoffs between number of stages, concentrations, and recovery.
- Catalyst addition process configuration studies.

(b) Gasification Recycle Gas Loop Research
  - Identification of preferred gasifier operating conditions.
  - Identification of technical uncertainties and data needs associated with trace impurities.
  - Evaluation of improved gas separation schemes.

6.3.5 Engineering Technology Studies

The following lists the engineering study efforts planned for the CCG process.

(a) Catalytic Gasifier Solids Balance Model
  - Modify proprietary computer model for use with CCG.
  - Validate model with available PDU data.

(b) Wet Scrubber Operability and Performance.
  - Evaluate interfacial properties of CCG solid-liquid streams.
  - Carry out lab studies on wet scrubber performance.

(c) Slurry Rheology and Solid-Liquid Separations for Catalyst Recovery.
  - Identify and evaluate solid-liquid separation alternatives.
  - Measure viscosity of char-catalyst slurries.

(d) Vapor-Liquid Equilibria in Sour Water/Catalyst Systems.
  - Review data needs to establish deficiencies
  - Undertake experimental program as needed to improve data base.

(e) Physical and Thermodynamic Properties of Catalyst Recovery Solutions.
  - Establish likely data needs.
  - Collect and evaluate available data.

(f) Dynamic Simulation of CCG Reactor System.
  - Develop dynamic process control
  - Determine response to changes in major variables.

(g) Environmental Control: Water and Solid Effluents.
  - Characterize and evaluate waste water streams from PDU.
  - Identify treatment alternatives.

(h) Environmental Control: Atmospheric Emissions.
  - Identify and quantify emissions through PDU testing.
  - Identify control alternatives.

(i) Preheat Furnace Tube Selection.
  - Evaluate carbonization behavior of selected tube materials.
  - Screen and select projected commercial furnace tube materials.

(j) Evaluation of Construction Materials for Catalytic Gasification.
  - Test and evaluate materials for CCG in PDU.

6.3.6. PDU Flow Diagram

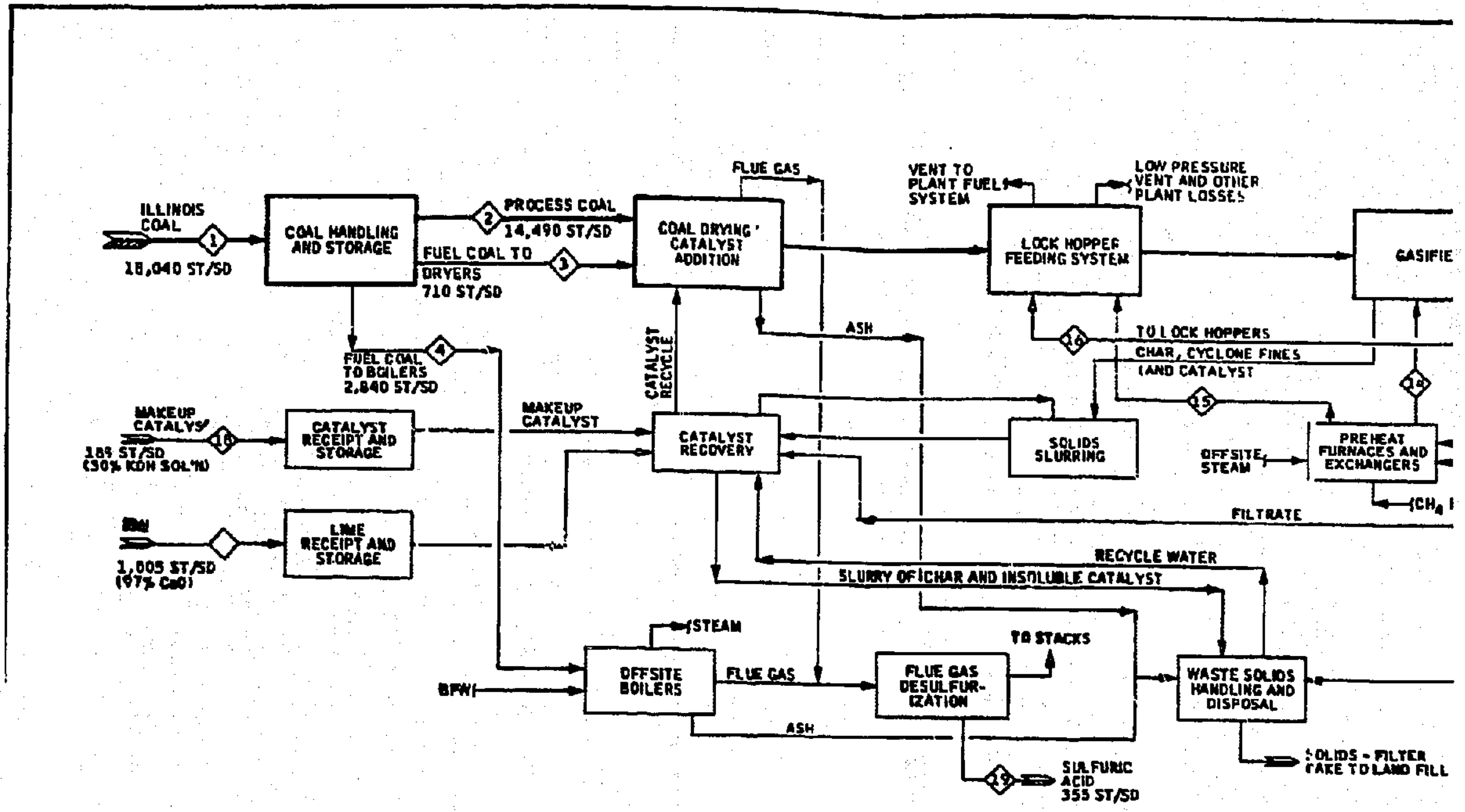
Included as Figures V-3 through V-5 are the PDU flow diagram, PDU gas separation section, and cryogenic methane separation scheme in the PDU, respectively.

7.0 REFERENCES

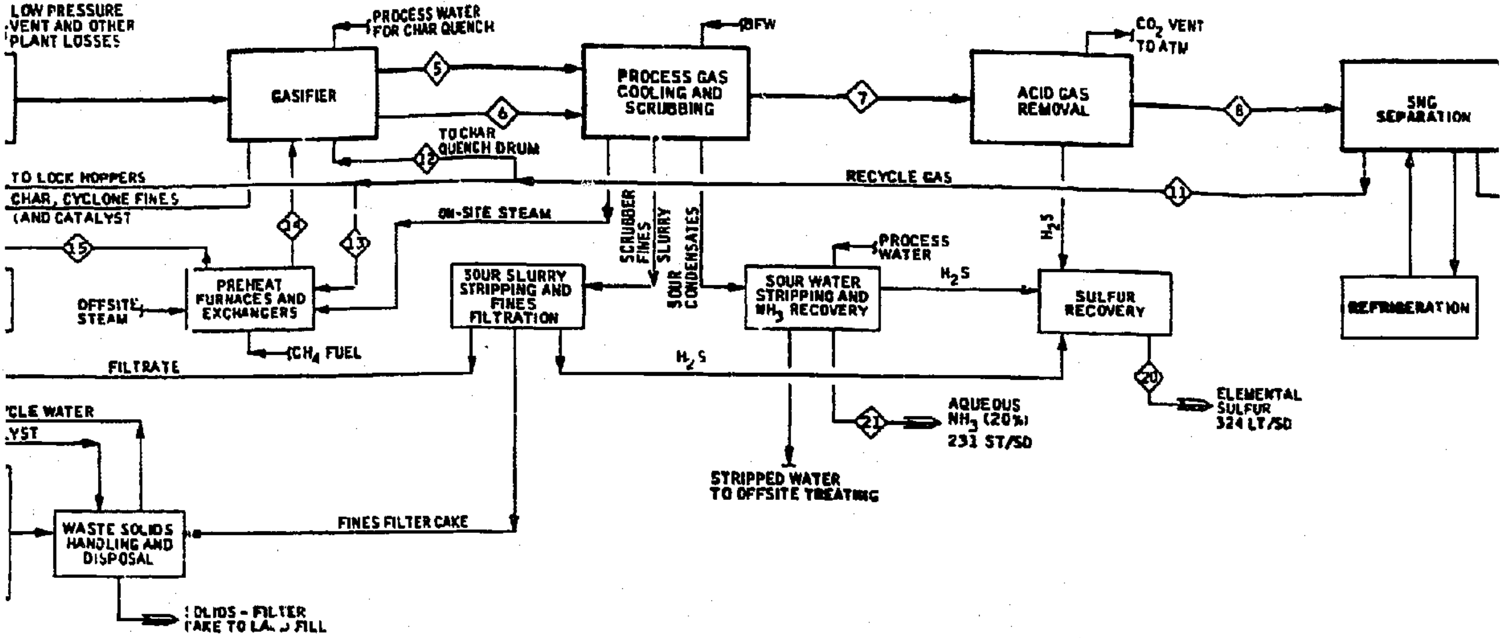
(1) Kalina, T. and Nahas, N. C., "Exxon Catalytic Coal Gasification Process - Predevelopment Program," Final Project Report, prepared by Exxon Research and Engineering Company for the U. S. DOE under Contract No. E(49-18)-2369, December 1978.

(2) Carpenter, D. J. and Thomas, D. A., "Low-Grade Refinery Heat Recovery Merits Attention," Oil and Gas Journal, January 28, 1980.

(3) Holdorff, G., "Revisions Up Absorption Refrigeration Efficiency," Hydrocarbon Processing, July 1979.



STREAM No.	1	2	3	4	5	6	7	8	9	10	
STREAM NAME	COAL FEED TO PLANT	COAL FEED TO PROCESS PLANT	FUEL COAL TO DRYERS	FUEL COAL TO BOILERS	GASIFIER PRODUCT GAS	CHAR QUENCH EFFLUENT	PROCESS GAS TO ACID GAS REMOVAL	TREATED GAS TO SNG SEPARATION	CH_4 FOR FUEL AND REFRIG MAKEUP	SNG PRODUCT	T RECY
COMPONENTS, lb-mol/hr											
CO	-	-	-	-	9,655	858	10,510	10 502	2	28	1(
CO_2	-	-	-	-	22,765	4	21,633	44	-	-	
H_2	-	-	-	-	34,597	3,082	37,671	37.665	-	-	37
CH_4	-	-	-	-	35,196	470	35,657	35 300	1,611	27,942	5
C_2H_6	-	-	-	-	5	-	5	3	-	3	
H_2S	-	-	-	-	947	-	878	.	-	-	
COS	-	-	-	-	14	-	2	.	-	-	
NH_3	-	-	-	-	1,180	-	-	.	-	-	
N_2	-	-	-	-	3,292	293	3,585	3.585	-	-	3
SUBTOTAL - DRYGAS	-	-	-	-	107,651	4,707	109,941	87,399	1,613	27,973	57,
H_2O	-	-	-	-	51,380	1,045	416	12	-	-	
TOTAL GAS, lb-mol/hr	-	-	-	-	159,031	5,752	110,357	87,411	1,613	27,973	57,
Mlbs/hr	-	-	-	-	2,978	65	2,033	1,039	26	449	
SOLIDS, M lbs/hr											
MAF (COAL, CHAR, etc)	-	-	-	-	-	-	-		-	-	
ASH (MINERAL MATTERS)	-	-	-	-	-	-	-	.	-	-	
TOTAL SOLIDS, Mlbs/hr	1,503	1,207	59	237	-	-	-	-	-	-	
TOTAL (GAS & SOLIDS), Mlbs/hr	1,503	1,207	59	237	2,978	65	2,003	1,039	26	449	
MMSCFD - GAS	-	-	-	-	1,448.45	52.39	1,005.13	793,41	14.69	254.78	
MOLE WEIGHT - GAS	-	-	-	-	18.73	11.30	18.42	11.93	16.06	16,06	
TEMPERATURE, °F					1,275	600	120	2	105	120	
PRESSURE, psia					500	500	450	430	45	1,015	



SEE NOTE (1)

	9	10	11	12	13	14	15	16	17	18	19	20	
ED GAS ING ATION	CH_4 FOR FUEL AND REFRIG MAKEUP	SNG PRODUCT	TOTAL RECYCLE GAS	RECYCLE GAS TO CHAR QUENCH DRUM	RECYCLE GAS TO PREHEATER	STEAM AND RECYCLE GAS TO GASIFIER	RECYCLE GAS TO FEED COAL	RECYCLE GAS TO LOCK HOPPERS	LIME FEED TO PLANT	CATALYST MAKEUP	SULFURIC ACID BYPRODUCT	RECOVERED ELEMENTAL SULFUR	AQU AMM
502	2	28	10,472	858	9,205	6,308	1,300	409	-	-	-	-	
44	-	-	44	4	37	1,629	5	3	-	-	-	-	
565	-	-	37,465	3,082	33,094	30,017	4,674	1,489	-	-	-	-	
500	1,611	27,942	5,747	470	5,050	4,337	713	227	-	-	-	-	
3	-	3	-	-	-	-	-	-	-	-	-	-	
	-	-	-	-	-	-	-	-	-	-	-	-	
	-	-	-	-	-	-	-	-	-	-	-	-	
	-	-	-	-	-	-	-	-	-	-	-	-	
85	-	-	3,585	293	3,150	2,705	445	142	-	-	-	-	
99	1,613	27,973	57,513	4,707	50,536	44,996	7,137	2,270	-	-	-	-	
12	-	-	12	1	11	86,008	2	-	-	-	-	-	
11	1,613	27,973	57,525	4,708	50,547	131,004	7,139	2,270	-	-	-	-	
39	26	449	564	64	496	2,004	70	22	-	-	-	-	
	-	-	-	-	-	-	-	-	-	-	-	-	
	-	-	-	-	-	-	-	-	-	-	-	-	
	-	-	-	-	-	-	-	-	84	16	-	-	
39	26	449	564	64	496	2,004	70	22	84	16	30	30	1
3.41	14.69	254.78	523.94	42.88	460.38	1,193.10	65.02	20.68	-	-	-	-	
11.93	16.06	16.06	9.81	13.63	9.81	15.30	9.80	9.78	-	-	-	-	
2	105	120	175	175	175	1,543	800	175					
0	45	1,015	600	520	600	520	555	555					

REVISIONS

LTR	ZONE	DESCRIPTION
A		INITIAL RELEASE

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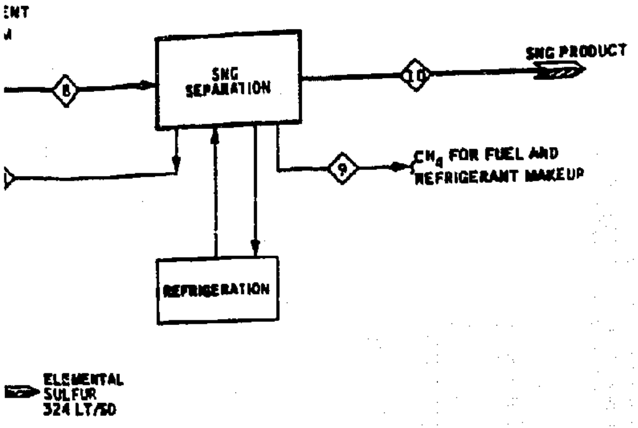

NOTE:

(1) 1,597 mol/hr REACTED BY WATER-GAS SHIFT REACTION IN THE PREHEAT FURNACES.

19	20	21
SULFURIC ACID BYPRODUCT	RECOVERED ELEMENTAL SULFUR	AQUEOUS AMMONIA
-	-	-
-	-	-
-	-	-
-	-	-
-	-	-
-	-	-
-	-	-
-	-	-
-	-	-
-	-	-
-	-	-
-	-	-
-	-	-
-	-	-
-	-	-
-	-	-
30	30	19
-	-	-
-	-	-

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PROCESS BLOCK FLOW DIAGRAM
EXXON CATALYTIC COAL
GASIFICATION PROCESS

SIZE	CODE IDENT NO.		REV
J	23835	FIGURE V-1	

SCALE | SHEET OF

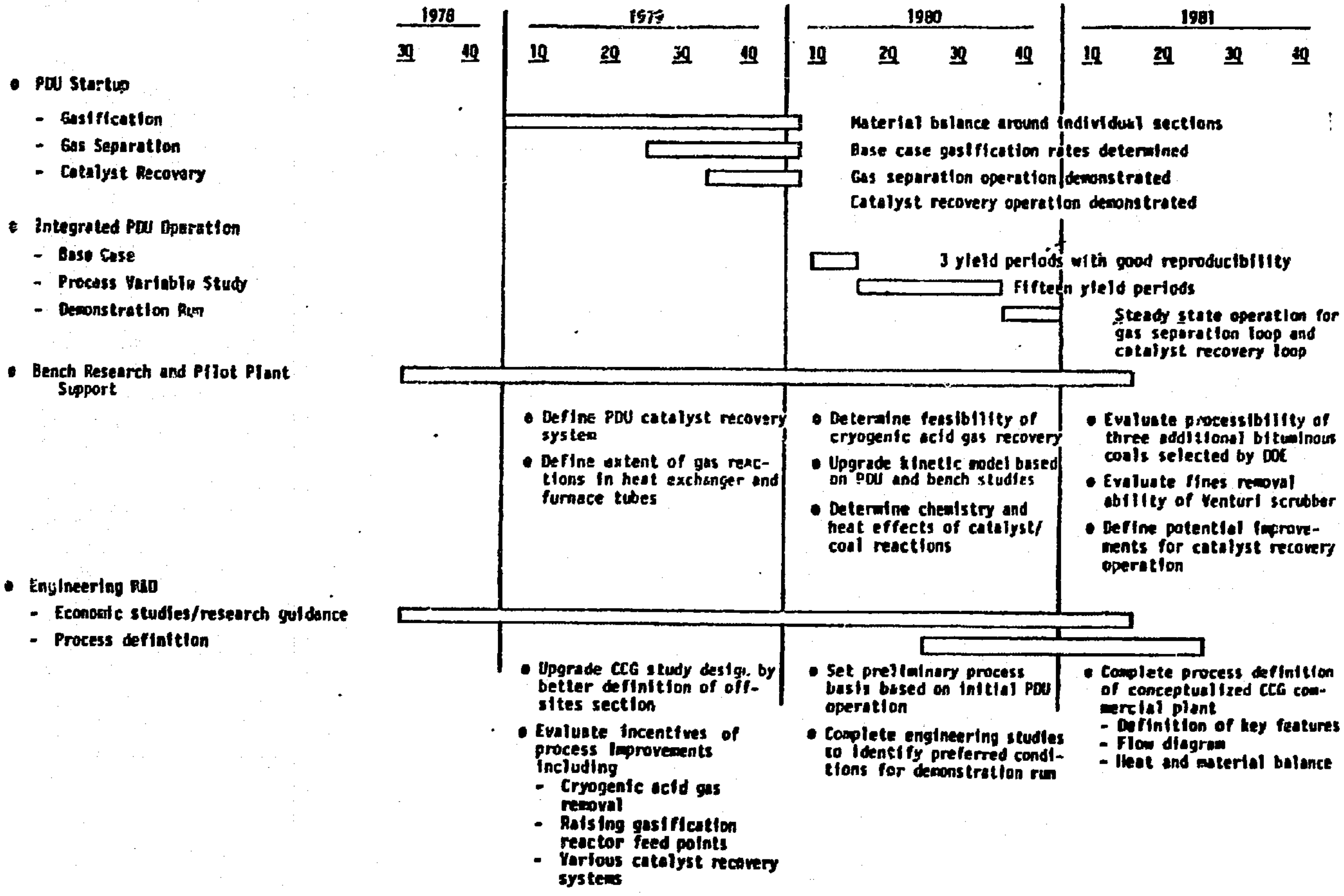


FIGURE V-2. Proposed Revised Schedule for SNG Program

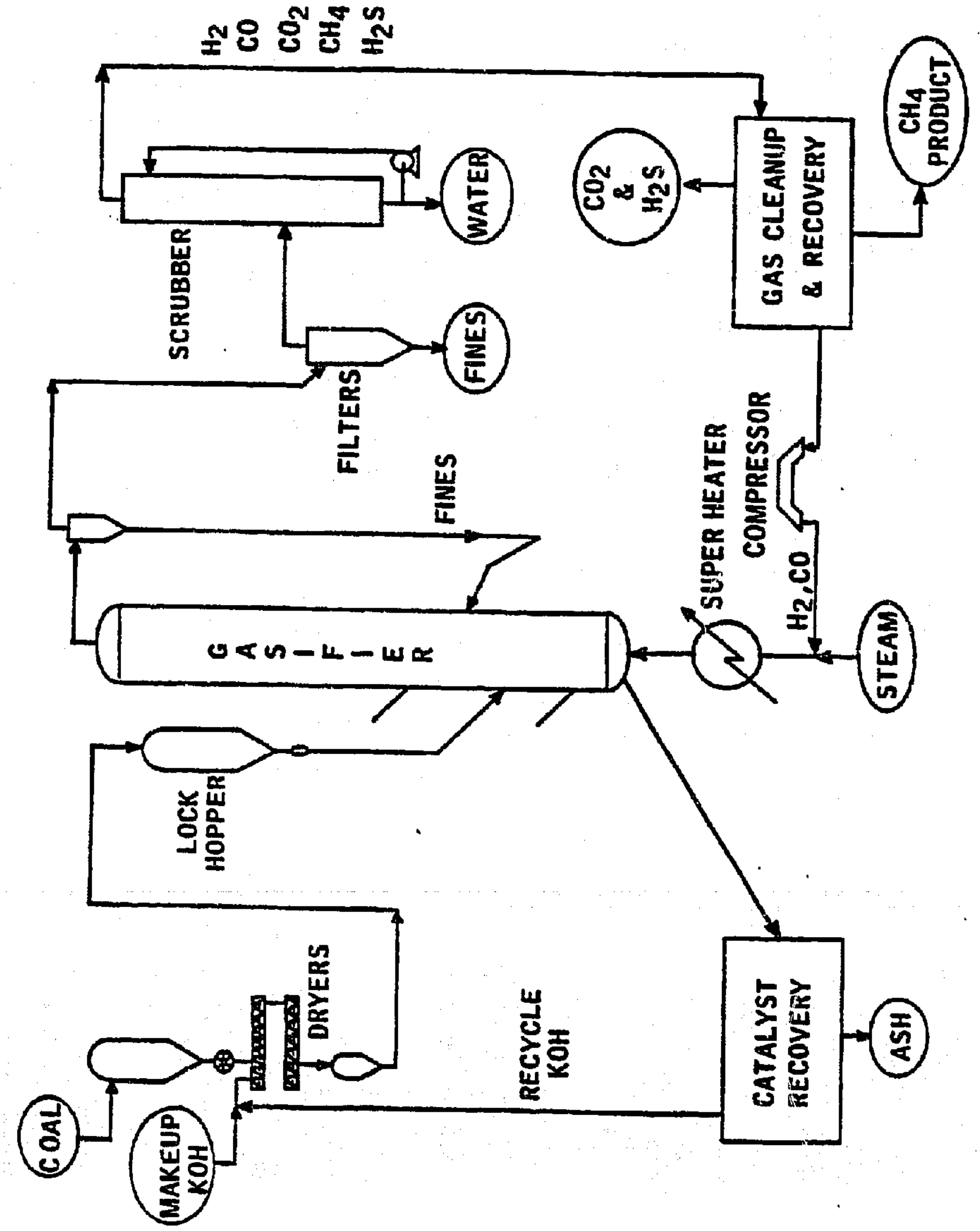
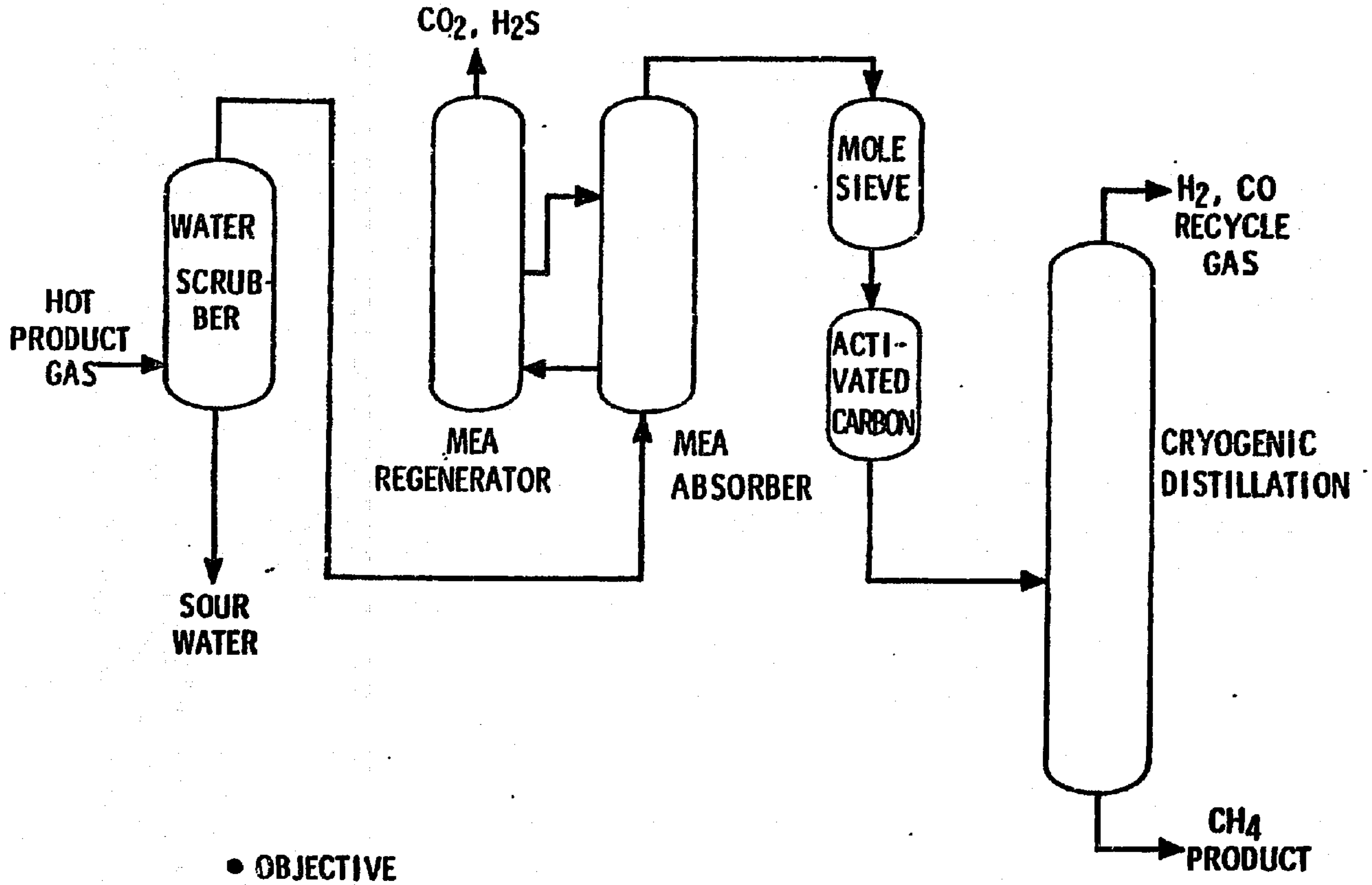
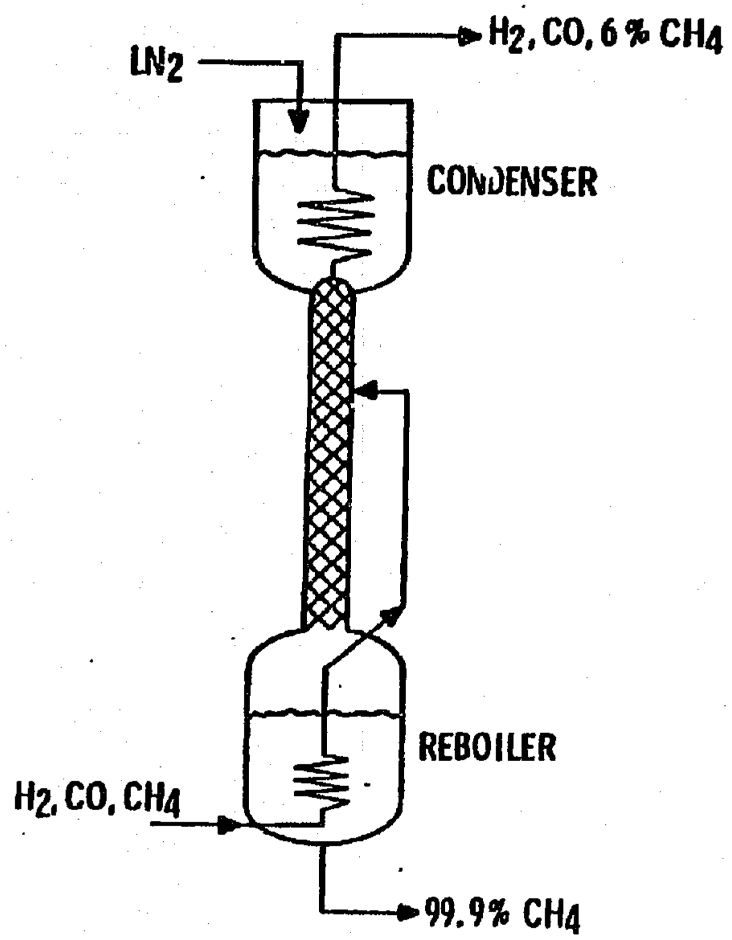


FIGURE V-3. Exxon CCG PDU Flow Diagram.



- OBJECTIVE
  - DEMONSTRATE OPERABILITY
  - MEASURE BUILDUP OF TRACE COMPONENTS

FIGURE V-4. Exxon CCG Gas Separation System



- SMALL UNIT TESTS SUCCESSFUL
  - SIMULATED FEED GAS
  - PRODUCT SPECIFICATIONS MET
  - DEMONSTRATED HEAT INTEGRATION
  - SMOOTH OPERATION
- BASIS FOR SETTING PROCESS DEVELOPMENT UNIT DESIGN

FIGURE V-5. Exxon CCG Cryogenic Distillation of Methane