EXXON CATALYTIC COAL GASIFICATION PROCESS PREDEVELOPMENT PROGRAM. FINAL PROJECT REPORT

EXXON RESEARCH AND ENGINEERING CO.
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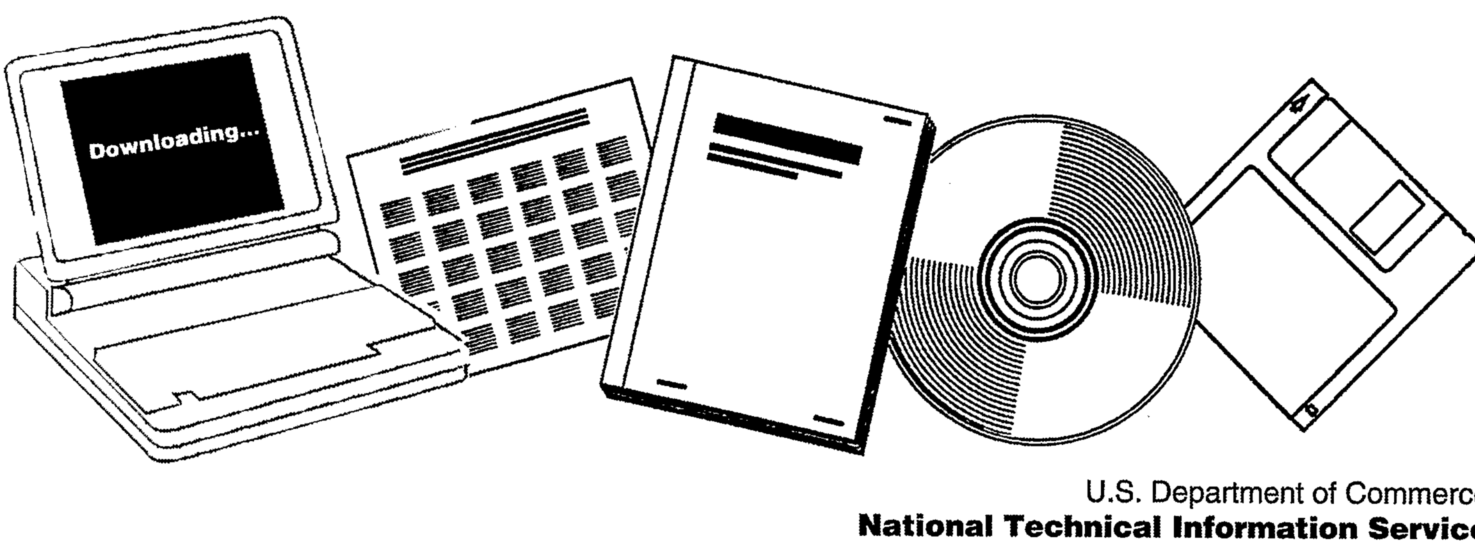

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EXXON CATALYTIC COAL GASIFICATION PROCESS PREDEVELOPMENT PROGRAM

FINAL PROJECT REPORT

T. Kalina
N. C. Nahas
Project Managers

EXXON RESEARCH AND ENGINEERING COMPANY
Baytown, Texas 77520

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ABSTRACT

This report summarizes the results of work conducted on Predevelopment Research for the Exxon Catalytic Coal Gasification Process. The eighteen-month effort (July 1976-December 1977) was a coordinated program which included operation of a continuous fluidized-bed gasifier, parallel bench-scale research, and engineering studies leading to the preparation of a commercial-scale plant study design and economics for producing SNG from Illinois coal.

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INTRODUCTION AND SUMMARY

This report covers the Predevelopment Program activities for the Exxon Catalytic Coal Gasification Process. The focus of the program was on the production of substitute natural gas (SNG) from bituminous coal. This work was performed by the Exxon Research and Engineering Company (ER&E) and supported by the United States Department of Energy under Contract No. E(49-18)-2369. The Predevelopment Program extended from July 1, 1976, through December 31, 1977.

The technical program during this predevelopment phase supports the transition to larger scale, more integrated testing in a development phase program. This section of the report first reviews the process description and results obtained prior to the predevelopment program. The objectives and results of the predevelopment program itself are then summarized.

Process Description

The Exxon Catalytic Coal Gasification Process combines the use of alkali metal salts as a gasification catalyst with a novel processing sequence which maximizes the benefits which can be derived from use of such a catalyst. The principal benefits from using an alkali metal gasification catalyst are that it increases the rate of steam gasification, reduces agglomeration of caking coals, and promotes the achievement of gas compositions closely approaching gas phase methanation equilibrium.

The process combines a relatively low gasifier temperature of about 1300°F with separation of synthesis gas ($CO + H_2$) from the product methane and recycle of the synthesis gas to the gasifier. Thus the only net products from gasification are CH_4, CO_2, and small quantities of H_2S and NH_3. The resulting overall gasification reaction can be represented as follows:

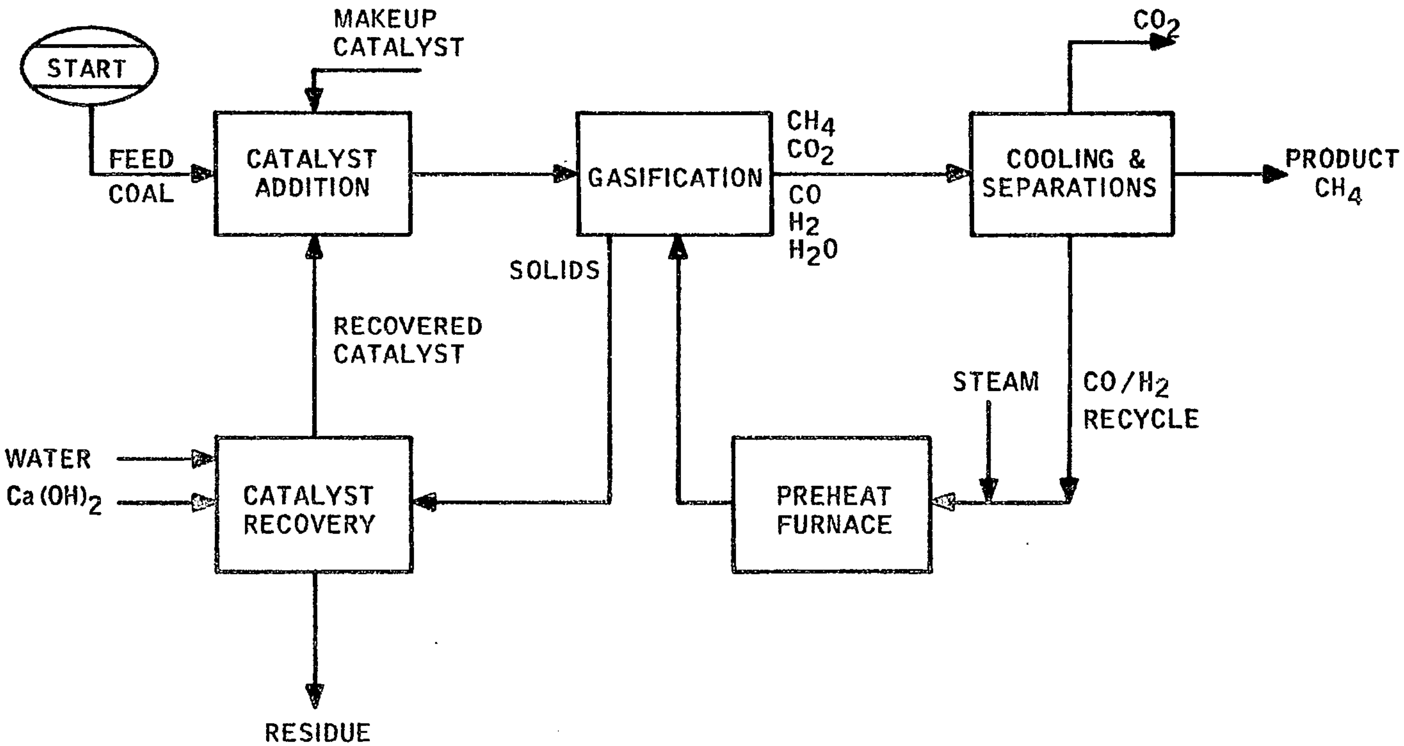
$$\text{Coal} + H_2O \rightarrow CH_4 + CO_2$$

Since this reaction is essentially thermoneutral, major heat input to the gasifier is not required.

A simplified flow plan for the Exxon Catalytic Coal Gasification Process is shown in Figure 0.1-1. Crushed coal is impregnated with catalyst, dried and fed via a lockhopper system to a fluidized bed gasifier which operates at about 1300°F and 500 psia. The coal is gasified with steam mixed with recycled synthesis gas, and the major gasifier product components are CH_4, CO_2, recycled CO and H_2, and unconverted steam. The catalyst reduces agglomeration of caking coals and no significant tars or oils are produced. Following heat recovery and water scrubbing, the product gas is treated in a series of separation steps including acid gas scrubbing to remove CO_2 and

FIGURE 0.1-1

EXXON CATALYTIC COAL GASIFICATION PROCESS



H_2S, and cryogenic fractionation to separate product methane from synthesis gas (H_2 and CO). The synthesis gas is combined with feed steam, preheated to approximately 250°F above the gasification temperature, and recycled to the gasifier. Although there is no net heat required for the gasification reaction, some heat input is required to heat up the feed coal, vaporize residual water, and provide for gasifier heat losses.

Solid residue from the gasification step contains about 10% of the coal carbon and all of the ash and catalyst. It is sent to a catalyst recovery step in which a large fraction of the catalyst is recovered from the residue using a calcium hydroxide digestion followed by countercurrent water washing. The recovered catalyst, along with some makeup catalyst, is added to fresh coal to complete the catalyst recovery loop.

Summary of Previous Research Results

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 synthesis gas makeup (simulated recycled) 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 deve a first-pass computer model of a fluid bed catalytic gasification reactor.

In the area of catalyst recovery, the effectiveness of 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 investments and operating costs.

Predevelopment Program Objectives

The Predevelopment Program work was divided into three major tasks. The key research objectives for each task are listed below.

Task I - Fluid Bed Gasifier (FBG) Studies With Illinois Coal

- Operate with mixed K_2CO_3/Na_2CO_3 catalyst
- Operate with recycled catalyst

Task II - Bench-Scale Studies

- 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

Task III - Engineering Research and Development

- Continue engineering screening studies
- Prepare an updated commercial plant study design

Summary of Predevelopment Program Results

The bench-scale research activities generated several significant results:

(1) A 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.

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

(3) Effort was redirected toward increasing the recovery level of the more effective potassium. This led to a preferred chemistry sequence capable of recovering more than 90% of the potassium catalyst.

(4) 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.

(5) 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.

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

The continuous fluid bed gasifier (FBG) was operated to simulate all commercial gasifier parameters except pressure, its effect on 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 FBG operations are summarized below:

(1) The unit was used to develop fifty material balanced periods. Of these, eighteen were selected to represent a variety of process variables for detailed work-up. Unit operations were of high quality. The service factor during the last six months of operation averaged more than 70% of real time, with a one-month maximum of 96%.

(2) FBG operations confirmed the ineffectiveness of mixed sodium and potassium catalyst.

(3) 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 build-up 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.

The engineering screening studies led to the following major conclusions:

(1) 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.

(2) 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.

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

(4) Selective heavy glycol scrubbing for acid gas removal is somewhat lower in cost than scrubbing with non-selective hot potassium carbonate or selective refrigerated methanol.

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:

(1) 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 1,640 million dollars (M$).

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.

(2) The estimated cost of SNG produced from this pioneer gasification plant is \$6.40 per million Btu (\$/MBtu). 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 \$4.80 per MBtu.

(3) Several factors could reduce the SNG cost below the Study Design range of \$4.80-6.40/MBtu. 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, which is currently in progress, will show a significant incentive for further development of the Catalytic Coal Gasification Process.