COLLECTED WORK NO. 24

SURVEY OF COAL LIQUEFACTION PRODUCTS INCLUDING SUITABILITY AS PETROCHEMICAL FEEDSTOCKS

J. B. O'HARA
N. E. JENTZ
J. E. PAPSO
THE RALPH M. PARSONS COMPANY

Presented at

AMERICAN INSTITUTE OF CHEMICAL ENGINEERS
SEVENTY-NINTH NATIONAL MEETING
HOUSTON, TEXAS

March 18, 1975

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The Ralph M. Parsons Company
Pasadena, California

1. INTRODUCTION

Events during the past decade affecting the world energy supply and demand balance foreshadowed justified concern by the petrochemical industry over future sources and compositions of feedstocks.

One industry that experienced major growth during this period was the petrochemical industry, which provided consumer products based on use of the building blocks ethylene, propylene, and the BTX mononuclear aromatics. As a result, an intensive effort began to find an assured and economical source of raw materials for production or separation of these compounds. Similar efforts were directed toward feedstocks for other petrochemicals.

The United States, western European, and Japanese ethylene and propylene industries developed with differing feedstock bases. The U.S. industry has been heavily oriented toward use of light aliphatic feedstocks, as represented by ethane and the LPG grouping. These materials were extracted from "wet" natural gas. For a time, supplies were adequate and costs attractive. The resulting ethylene plants were simpler and less expensive than those based on heavier feedstocks, and there was a limited by-product sales/disposition requirement. The plants were heavily concentrated in geographical areas close to the feedstock supply. Western European and Japanese petrochemical industries, however, were not blessed with comparable indigenous LPG supply. They favored use of naphtha feedstocks.

The U.S. energy balance, as well as LPG supply/demand, has heavily impacted the petrochemical feedstock situation with respect to assured supply and operating costs for existing plants, and also for planning for future plants. The situation is that the LPG supply depends heavily on "wet" natural gas supply sources, which have been declining in recent years. This decline is expected to continue unless current trends are reversed by the discovery and development of significant new reserves.

Turning our attention now to a broader look at the future feedstock supply question for the petrochemical/chemical industries, and the role that a possible future coal conversion industry might play in feedstocks, we note that, during its formative years, the world's organic chemical industry was largely based on the use of coal as the basic raw material. Using coal, it successfully supplied the markets for products such as methanol, phenol, and polymers as illustrated by synthetic rubber. Ammonia and ammonium sulfate represent demonstrated commercial inorganic products made from coal.

Specific examples of historical coal-based industries are the German dye industry since 1880; calcium carbide and acetylene from coke (1896); the DuPont ammonia-methanol plant at Belle, West Virginia (1927) [1]; numerous coal tar distilleries; and the Union Carbide coal hydrogenation plant at Institute, West Virginia, which is reported to have operated from 1950 to 1957.

Coal represents the largest U.S. reserve of fossil energy. It is expected to supply a larger share of our indigenous energy supply during the coming years. The simplest way to use coal is to burn it!

However, there are incentives to convert it to ecologically "clean" liquid, gas, or solid forms. One incentive is to meet environmental standards. Another is to put it in a more convenient and valuable form for distribution and storage. Still another would be to convert it to feedstocks for use in existing and planned petrochemical plants. The incentives must be significant enough to justify the conversion because it is expensive and difficult to convert coal to gas or liquid.

The executive branch of the government has stated that the development of a viable coal conversion industry is a national priority objective [2]. A specific near-term objective is production of the equivalent of a million barrels per day of synthetic fuels from coal by 1985. Plans to achieve this objective are being developed and evaluated. Discussions regarding a parallel objective - namely, the potential for supply of petrochemical feedstocks from coal - are, therefore, timely; and merging of planning efforts and exchange of views on objectives by the coal conversion and petrochemical communities have the promise of benefiting both.

Based on the experience of The Ralph M. Parsons Company in the roles of Technical Evaluation Contractor for the field of clean liquid and/or solid fuels from coal and supplier of Preliminary Design Services to the Office of Coal Research, this paper will briefly summarize views on expected future feedstock requirements by the petrochemical industry and their potential supply from a coal base. Projections will be made regarding potential coal-derived feedstocks for existing plants and also possibilities of design considerations for future petrochemical plants to utilize products of the coal conversion industry. Maximization of future use of coal-derived products may also affect industry and government R&D program plans.

2. OBJECTIVE

The primary objective of this paper is to present a preliminary assessment of the contribution that a coal conversion industry might make to the projected future feedstock supply pool for the petrochemical industry.

3. PETROCHEMICAL FEEDSTOCK DEMAND

3.1 HISTORICAL

The year 1955 has been arbitrarily selected as the initial reference date for historical data. This year roughtly coincided with an increased level of activity in the industry.

Figure 1 [3] shows the historical production for the two high-volume olefins, ethylene and propylene, as well as BTX (benene, toluene, and xylene). It also shows projections to 1985. The five compounds referred to as primary petrochemicals constitute more than half of the weight of the total petrochemical production [4,5].

For the period 1973-1975, the petrochemical industry feedstock consumption is expected to amount to approximately 6% of the total U.S. refined product output [6]. Including feedstock and plant fuel requirements, the industry is expected to use about 10% of the total natural gas supply. While these are modest percentages of the liquid and gas supplies, they support an industry that multiplies their product value significantly.

Figure 2 shows the estimated ethylene feedstock requirements [7]. For 1974, ethane and LPG supplied about 75% of the total; their share is projected to drop to approximately 45% by 1980, with naphtha and gas oil usage increasing to make up the difference. Further shifts in the direction of use of heavier feedstocks are indicated unless competitive light feedstock supplies can be developed.

For comparison, Figure 3 shows the amounts of ethane and LPG that the industry could use to produce the quantity of ethylene projected in Figure 1 if the feedstocks continued to be available to permit retention of the same proportions used in 1974; also shown is the projected actual feedstock mix and the shortfall in the desired light feedstocks. This shortfall of about 250,000 and 300,000 B/D for ethane and LPG, respectively, in 1985 provides a target for the synthetic fuels industry to determine its ability to competitively supply this market. A similar target exists for propylene feedstocks.

4. POTENTIAL TYPES OF FEEDSTOCK SUPPLY FROM COAL LIQUEFACTION OPERATIONS

4.1 EXAMPLE COMPOSITIONS OF COAL-DERIVED LIQUIDS

A number of processes now under development are intended to produce liquid and clean solid fuels from coal. While plans are underway to determine the detailed chemical compositions of the liquids, this phase of the work is in a very early stage of development. An example is the Dow Chemical project to analyze coal-derived liquids and then prepare a feasibility study for production of chemicals from coal liquefaction products; this work is being done under Contract No. 14-32-0001-1534 for the Energy Research and Development Administration (ERDA). The summary to be presented here must, therefore, be considered only as a starting point. It is planned that it will be updated in the future as additional information becomes available.

The following paragraphs will discuss sample compositions of coal-derived liquids from at least one process for each category of coal liquefaction processes. For convenience, coal liquefaction processes can be classified as hydroliquefaction, pyrolysis, extraction, and indirect; Fischer-Tropsch technology illustrates indirect liquefaction.

An example of an inspection of a hydroliquefaction product is given in Figure 4 [8]. This combination results in the following weight percent yields of hydrocarbon products; the solvent is recycled to the conversion system:

Product	Yield (Wt. %)
C_1 - C_3 Range	5.9
C_4 - C_6 Range	2.8
"Light" Hydrocarbons	4.4
Solvent Refined Coal (SRC)	54.7
Total, Wt. % of Feed Coal	67.8

Compounds that have been identified in the lower boiling regions are listed in Figure 5 [9]. A modest amount of aliphatics, including paraffins, are produced.

A sample product yield structure for a catalytic hydroliquefaction process is shown in Figure 6 [10]. The C_2 and C_3 components represent approximately 8% of the yield as shown. Additional details are shown in Figures 7 and 8 [11]. The complexity of the product mixture is apparent from inspection of these data. The light aliphatics and aromatics are of particular interest.

Figure 9 [12] illustrates the gas composition produced in the COED pyrolysis process. Hydrocarbons in the C_2 - C_4^+ range in this case represent 14.5% of the gas produced. For a 25,000-ton-per-day coal feed plant, the hydrocarbon gases/vapors production would be of the order of 2,500 tons per day.

Examples of raw and hydrotreated COED oil are shown in Figure 10. Approximately 40% of the raw oil, recovered from the vapor phase produced in the pyrolysis step, would boil below and 800°F gas oil range. Because of its aromatic nature, one of the potential uses of COED oil is in furnace black manufacture.

Figure 11 presents physical characteristics of a coal-derived synthetic fuel produced by an extraction process at the Office of Coal Research's Cresap, West Virginia, pilot plant [13]. This product had been hydrotreated to produce a 21°API low-sulfur synthetic fuel. Approximately 25% would be in the 350°F endpoint naphtha range. Additional data are required regarding its chemical composition in order to define its application as a chemical feedstock.

A sample product yield distribution from a fluidized-bed indirect or Fischer-Tropsch synthesis reaction at SASOL is given in Figure 12 [14]. The majority of the liquid product is in the gasoline range. Because of the nature of the synthesis reaction, paraffins will be formed.

To summarize, a characteristic of coal-derived liquids produced by hydroliquefaction, pyrolysis, and extraction is their aromatic, naphthene, and cycloparaffin content, resulting in differences in characteristics from petroleum-sourced liquids of similar boiling points. Liquids produced by Fischer-Tropsch technology tend to be more aliphatic in composition.

4.2 POTENTIAL USES OF COAL LIQUIDS IN THE PETROCHEMICAL AND CHEMICAL INDUSTRIES

Having developed some background of characteristics of coal-derived liquids, let's consider a few potential uses for them in the petrochemical and chemical industries. This portion of the discussion will be directed to products produced through processes other than the Fischer-Tropsch.

The possibilities of generation of some material for use in steam-cracking operations to produce olefins and as precursors for

BTX production has been mentioned earlier, and will be touched on again later.

Coal-derived liquids should be useful as a feedstock to produce needle coke. The heteratom content should be reduced prior to coke production; the sulfur content should be reduced to the range of 0.1 to 0.6 wt. %. A catalytic hydrogenation process should be used that does not hydrocrack or saturate the aromatics. This potential use deserves consideration because of the amount of processing required to produce equivalent feedstock from petroleum, whereas it appears that the coal-derived liquids would only require sulfur adjustment because the coal-derived liquids naturally possess an aromatic content equal to or greater than that in the feedstocks used to produce needle coke to date.

Coal-derived liquids are also well suited for use as feedstocks for carbon black or furnace black manufacture because of their highly aromatic nature. Evidence indicates that the coal-derived liquids can be used directly for this purpose.

A number of chemicals potentially can be recovered from the liquid products; examples are phenol, xylenol, cresylic acids, aniline, toluidine, xylidene, quinoline, and napthalene. United States Steel Corporation has an ERDA-sponsored program underway that will include the investigation of chemicals recovery from coal liquids.

Hydrocracking of the heavy oils followed by catalytic reforming and extraction should produce BTX and a paraffinic raffinate useful as a feedstock for steam-cracking operations. Process conditions should be selected to minimize the production of isoparaffins. Other potential sources of steam-cracker feedstocks are recovery from the vapor phases of the liquefaction and hydrotreating operations for the hydroliquefaction, pyrolysis, and extraction routes plus the synthesis product from Fischer-Tropsch plants.

Potentially, processing and refining of raw coal-derived liquids could be done either at the coal liquefaction or at the user's plant. There are incentives to carry them out at the coal conversion plant for greater efficiency and convenience at that plant, and then shipping the feedstock to the petrochemical or chemical plant for subsequent conversion, competing with or replacing petroleum-sourced raw materials.

5. POSSIBLE PRODUCTION QUANTITIES OF LIQUIDS FROM COAL

Available information does not permit accurate prediction of the number of coal liquefaction plants nor the quantity of coal-derived liquids that will be constructed over the next 20-year period. Political, economic, technological, and environmental factors will all contribute to the final decisions. For example, scenarios developed a year ago must be reevaluated in light of recent economic and political events.

One possible scenario is that the incentives to produce alternate petrochemical feedstocks from coal would lead to construction and operation of the number of plants and capacity as shown in Figure 13. This scenario indicates a total process coal feed rate of 100,000 tons per day of process coal in 1985, increasing to approximately 570,000 in 1995. Note that Figure 13 is presented as one possibility for use in illustrating the potential, and is not a forecast.

6. POTENTIAL PRODUCTION OF PETROCHEMICAL FEEDSTOCKS FROM COAL

To pursue the logic pattern to the point of developing a basis to judge possible implications of a feedstock-from-coal industry, a conjectural block flow diagram and material balance were developed for a coal conversion liquid-processing complex. The intent is to provide some starting basis for discussion regarding the character of a coal conversion complex keyed to interfacing with the petrochemical and chemical industries.

The schematic diagram depicting a possible combination of process steps is shown in Figure 14. The process steps shown, in addition to those for a sample hydroliquefaction plant, include:

(1) Coking

(2) Hydrocracking

(3) Hydrogen desulfurization

(4) Catalytic naphtha reforming

(5) Aromatics extraction

(6) Extraction and fractionation (phenols, tar acids, and amino compound production)

(7) Gas recovery and fractionation

The processing sequence is intended to capitalize on the aromatic nature of the feed material. Figure 15 shows projected quantities and compositions of chemical feedstocks that might be generated from coal-derived liquid. The quantities listed are percent of the total product from the feedstock generation complex. The "process" coal feed represents the feed to the coal conversion operation; additional energy is required to operate the complex.

It is worthwhile to restate that the array of chemical manufacturing feedstocks listed in Figure 14 represents only one of many possible arrays. The emphasis for the array shown has been placed upon high sales-volume products. One of the products selected could be coke production, which may produce enough to affect the existing market; the plan shown could produce as much as 20% of the coke demand. Where such might prove to be the case, technology exists to reduce the volume of coke manufacture.

The availability of feedstocks from the complex schematically depicted in Figure 14 and based on the total coal processing scenario shown in Figure 13 is given in Figure 16. Here we see that the production quantities from the coal processing plant over the period

1985 to 1995 could range from 20 to 115% of 1974 ethylene feedstock usage for propane-plus-LPG. For naphtha, the range could be 82 to 470%. Benzene production could range from 9 to 50% of 1974 production.

7. SUMMARY AND CONCLUSIONS

A portion of total desired petrochemical feedstocks could be produced from coal liquefaction operations if a number of coal liquefaction plants are constructed and operated. The value of the coal liquids to the feedstock user can be maximized by selection of recovery and processing steps used in the coal liquids processing plant. Additional information is required regarding the composition and characteristics of coal-derived liquids. Some work is under way with this objective in view.

The potential for petrochemical/chemical use of coal liquefaction should be further evaluated as the coal conversion development program progresses, and as sources of supply of feedstocks from classical sources change.

The economics of feedstocks-from-coal should be studied in detail.

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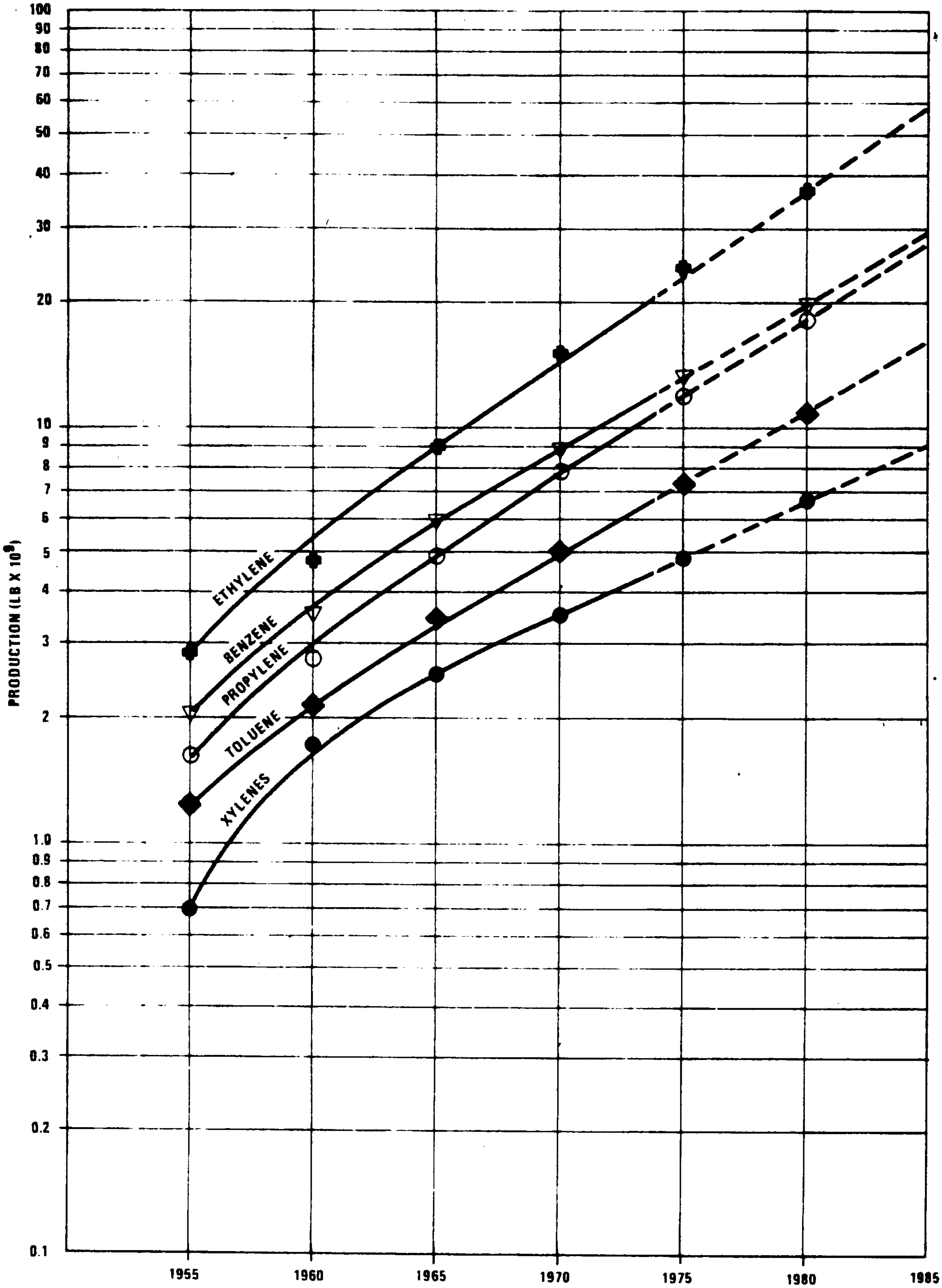


FIGURE 1 · U. S. PRIMARY PETROCHEMICALS PRODUCTION

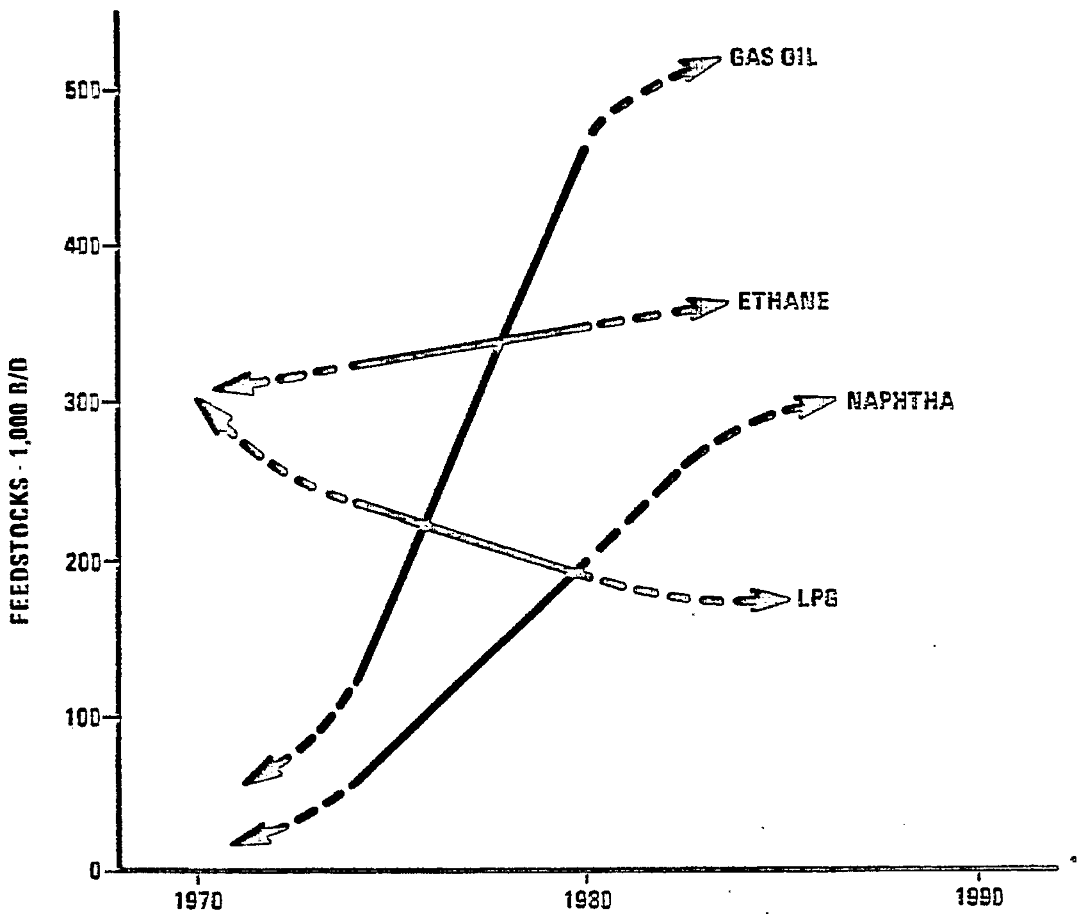


FIGURE 2 - ETHYLENE PLANT FEEDSTOCK CONSUMPTION

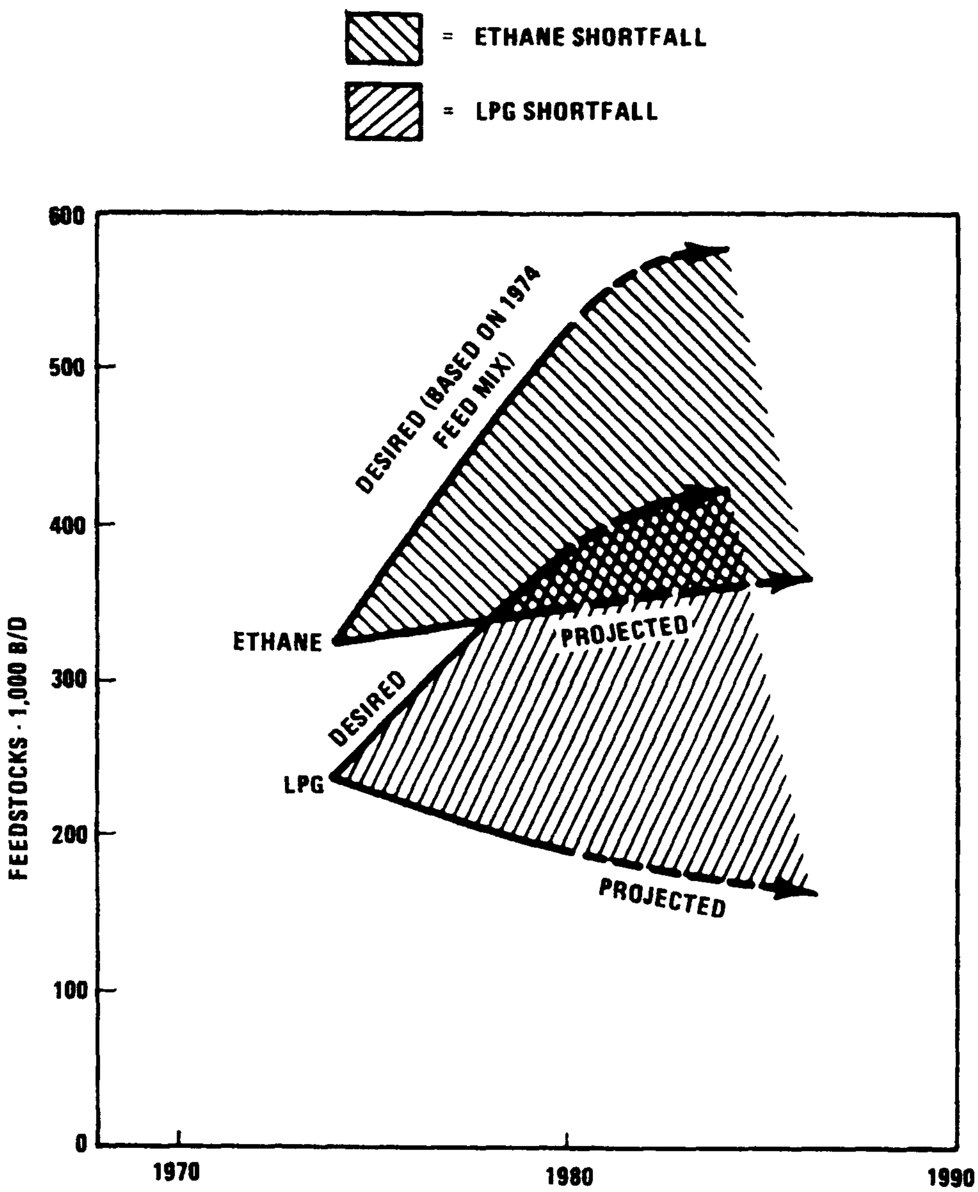


FIGURE 3 - PROJECTED ETHYLENE FEEDSTOCK AVAILABILITY TARGETS AND SHORTFALLS

FEED

COAL	100.0 POUNDS
H_2 CONSUMPTION	2.1
TOTAL	102.1

PRODUCTS	WEIGHT % OF COAL FED	°API	BOILING RANGE @ ATMOSPHERIC PRESSURE
C_1-C_3 RANGE	5.8		
C_4-C_5 RANGE	2.5		
LIGHT HYDROCARBONS	4.3	25.8	136 - 642°F
NET SOLVENT	10.3	0.3	373 - 975°F
SRC	53.6	-18	975°F +
UNDISSOLVED COAL	5.8		
ASH	10.9		
NET H_2O	6.2		
CO_2	1.1		
H_2S	1.5		
NH_3	0.1		
TOTAL	102.1		

FIGURE 4 - EXAMPLE: SOLVENT REFINED COAL (SRC) YIELD DISTRIBUTION

BOILING RANGE, °F	IBP-180	180-400	400-650
WT % OF DRY COAL	0.62	3.38	12.42
GRAVITY, °API	57.8	27.6	9.8
GAS CHROMATOGRAPH ANALYSIS, WT %			
C_3	0.73		
ISOBUTANE	1.52		
N-BUTANE	4.09		
ISOPENTANE	7.50		
N-PENTANE	3.76		
2-METHYLPENTANE	10.72		
3-METHYLPENTANE	12.99		
N-HEXANE	22.72		
METHYLCYCLOPENTANE	9.92		
BENZENE	11.20		
CYCLOHEXANE	12.73		
HEPTANES	2.13		
HYDROCARBON TYPES, VOL %			
MASS SPECTROMETER			
PARAFFINS		14.09	2.79
CYCLOPARAFFINS			6.79
ALKYLBENZENES			14.31
INDANES			22.90
INDENES			12.51
NAPHTHALENES			11.67
ACETYLNAPHTHENES			12.65
ACETYLNAPHTHALENES			11.09
TRICYCLOPARAFFINS			5.46
DICYCLOPARAFFINS		10.62	
TOTAL NAPHTHENES		45.24	
TOTAL AROMATICS		28.85	
AROMATIC DISTRIBUTION			
BENZENE		2.87	
TOLUENE		3.21	
C_8 AROMATICS		8.38	
C_9 AROMATICS		6.66	
C_{10} AROMATICS		5.10	
C_{11} AROMATICS		2.20	
C_{11} AROMATICS		0.43	
FIA ANALYSIS			
SATURATES		66.40	
OLEFINS		1.20	
AROMATICS		32.40	

FIGURE 5 - EXAMPLE: SOLVENT REFINED COAL (SRC)
COMPONENT ASSAY

NORMALIZED PRODUCT DISTRIBUTION	WT %
C_1-C_3 HYDROCARBONS	10.7
C_4-400°F DISTILLATE	17.2
400-650°F DISTILLATE	28.2
650-975°F DISTILLATE	18.6
975°F + RESIDUAL OIL	10.0
UNREACTED ASH-FREE COAL	5.2
H_2O, NH_3, H_2S, CO, CO_2	15.0
TOTAL (100.0 + H_2 REACTED)	104.9
COAL CONVERSION, %	94.8
HYDROGEN CONSUMPTION, SCF/TON OF PROCESS COAL	18,600

FIGURE 6 - EXAMPLE: PRODUCT YIELD DISTRIBUTION
H-COAL PROCESS

SATURATED COMPOUNDS	C_4-400°F FRACTION	400-650°F FRACTION	650-919°F FRACTION
COMPONENT	(WT %)		
nC_4	0.10		
iC_5	0.20		
nC_5	0.69		
C_6	2.48		
C_7	2.87		
C_8	2.08		
C_9	1.59		
C_{10}	1.19		
C_{11}	0.60		
C_{12}	0.11		
	11.99		
SATURATED NAPHTHENES	(WT %)	(WT %)	(WT %)
N-PARAFFINS		4.8	
I-PARAFFINS		1.7	1.4
MONOCYCLOPARAFFINS	42.64	14.0	3.1
BICYCLOPARAFFINS	8.50	7.9	0.6
TRICYCLOPARAFFINS	0.19	2.6	0.7
TETRACYCLOPARAFFINS			0.4
PENTACYCLOPARAFFINS			0.2
HEXACYCLOPARAFFINS			0.1
PHENYLS			0.3
	51.33	31.0	6.8
UNSATURATED NONAROMATIC COMPONENT			
MONOCYCLOPARAFFINS	5.32	4.3	0.5
BICYCLOPARAFFINS	4.98		0.3
TRICYCLOPARAFFINS	0.90		0.2
TETRACYCLOPARAFFINS			0.2
PENTACYCLOPARAFFINS			0.1
HEXACYCLOPARAFFINS			0.1
PHENYLS			0.2
	11.20	4.3	1.6

FIGURE 7 - EXAMPLE: COMPONENT DISTRIBUTION FROM H-COAL PROCESS

AROMATIC COMPOUNDS	C_4-400°F FRACTION	400-650°F FRACTION	650-919°F FRACTION
COMPONENT	WT %	WT %	WT %
ALKYL BENZENES		12.6	3.0
C_6	0.89		
C_7	3.77		
C_8	4.76		
C_9	4.16		
C_{10}	2.58		
C_{11}	1.29		
C_{12}	0.10		
	17.55	12.6	3.0
OTHER COMPOUNDS	WT %	WT %	WT %
INDSANS AND TETRALINS	6.44	30.8	0.5
INDENED		5.7	
NAPHTHALENE		0.2	
NAPHTHALENES	0.59	3.5	
ACENAPHTHENES (C_nH_{2n-14})		4.0	
ACENAPHTHENES (C_nH_{2n-16})		2.2	
TRICYCLICES (C_nH_{2n-18})		0.4	
OTHER AROMATICS			12.8
PHENOLS (MW)			1.5
108	0.13	0.04	
122	0.56	0.52	
136	0.19	0.98	
150	0.02	0.38	
164		0.07	
178		0.01	
OTHER NONHYDROCARBONS		3.10	13.8
	7.93	50.10	88.6
TOTAL WT %	100.00	100.00	100.00

FIGURE 8 - EXAMPLE: COMPONENT DISTRIBUTION FROM H-COAL PROCESS

PRODUCT	GAS COMPOSITION (DRY WT %)	DRY AND ACID GAS FREE (WT %)
H_2	3.19	5.45
N_2	0.79	1.35
H_2S	3.16	
CO_2	38.26	
CO	31.40	53.60
CH_4	8.70	14.85
C_2H_4	0.59	1.01
C_2H_6	1.53	2.61
C_3H_6	0.53	0.90
C_3H_8	0.29	0.50
C_4^+	11.56	19.73
	100.00	100.00
POUNDS GAS / POUNDS MF COAL	0.72	0.42

FIGURE 9 - EXAMPLE: COED PRODUCT GAS YIELD DISTRIBUTION

ASTM DISTILLATION VOL %	RAW COED OIL °F	HYDROGENATED COED OIL °F
IBP	385	108
10	500	245
30	720	356
50	870	465
60	960	515
69	1,015	–
70	-	559
90	-	673
EP	-	746
REC	-	98%
°API	−4	55
POUR POINT, °F	123	–
FLASH POINT, °F	392	ROOM TEMPERATURE 19,000
GROSS HEATING VALUE, BTU/LB	≃ 15,700	≃ 19,000
(POUNDS OIL / POUND MF COAL)	0.20	0.20 - 0.18

FIGURE 10 - EXAMPLE: COED OIL CHARACTERISTICS

ASTM DISTILLATION	TEMPERATURE °F
IBP	202
5%	250
10%	288
30%	396
50%	447
70%	492
90%	562
95%	598
END POINT	652
% OVER	98.0
% BOTTOMS	1.5
API AT 60°F	21.0
SP GR AT 60°F	0.928
S, WT %	0.0925
N, WT %	0.17
BROMINE NO	7
FIA A	75.3
O	0.9
P+N	23.8

FIGURE 11 - EXAMPLE: CHARACTERISTICS OF LIQUID PRODUCTS FROM EXTRACTION PROCESS

PRODUCT	WT %
LPG	0.6
GASOLINE	84.9
DIESEL OIL	3.7
WAXY OIL	1.2
METHANOL	0.4
ETHANOL (CRUDE)	0.3
ETHANOL (PURE, ANHYDROUS)	7.9
METHYL ETHYL KETONE	0.6
ACETONE (PURE)	0.4
	100.0

FIGURE 12 - EXAMPLE: FISCHER-TROPSCH YIELD DISTRIBUTION
FLUID BED SYNTHESIS OPERATION AT SASOL

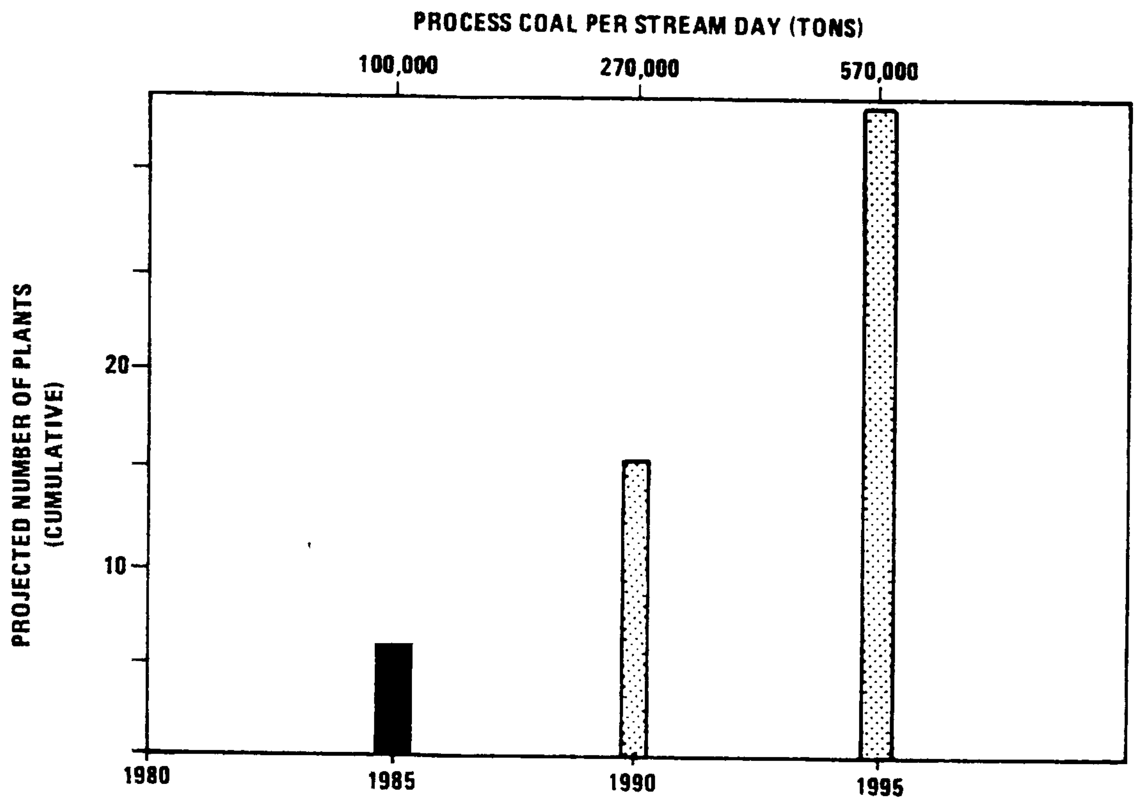


FIGURE 13 - POTENTIAL LIQUIDS FROM COAL LIQUEFACTION PLANTS

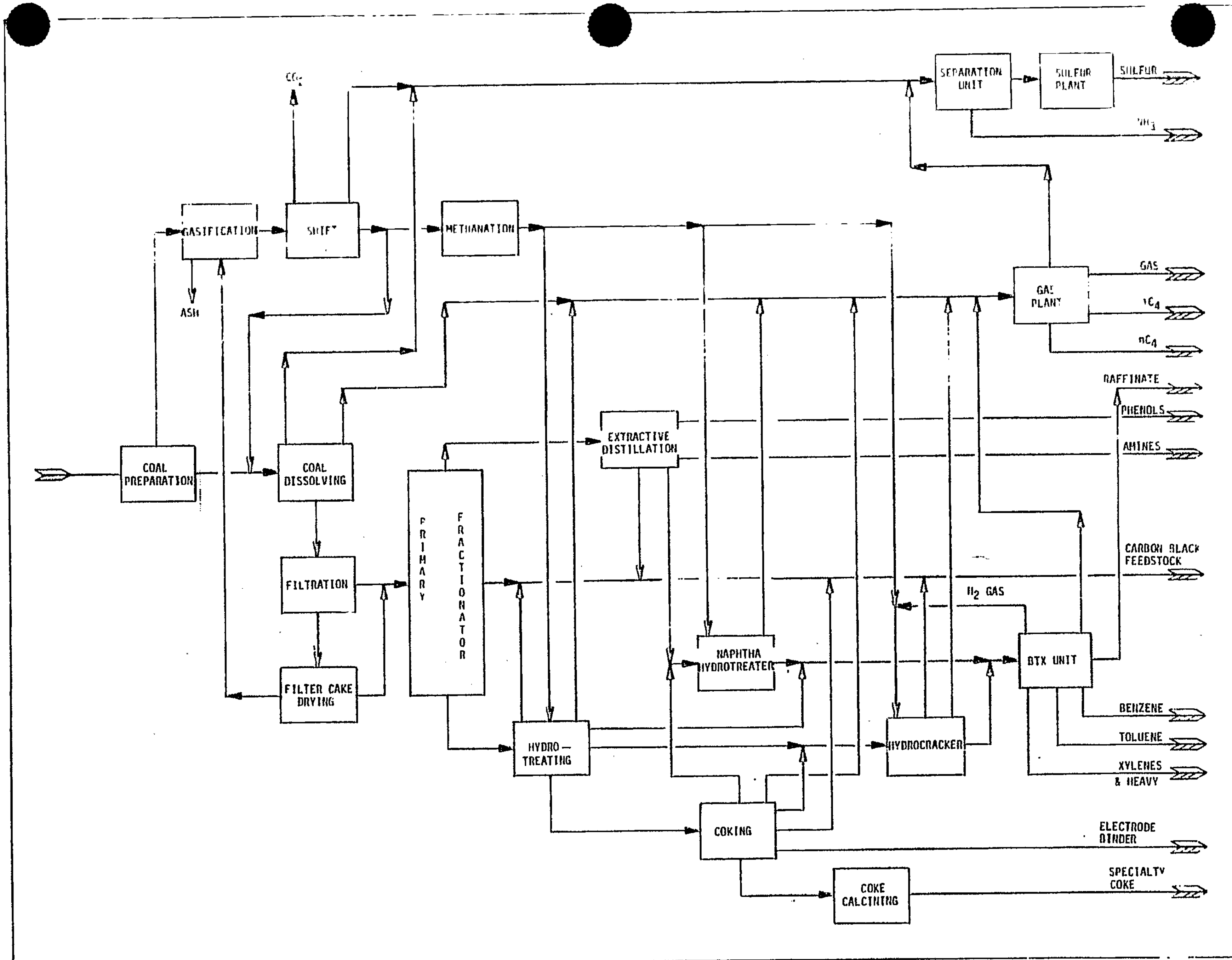


FIGURE 14 - BLOCK FLOW DIAGRAM; A POSSIBLE PROCESS SEQUENCE FOR PETROCHEMICAL FEEDSTOCK PRODUCTION

COMPONENT	PRODUCTION RATE, T/D BASIS 10,000 T/D PROCESS COAL FEED	PRODUCT COMPOSITION (WT %)
METHANE	476	6.1
ETHANE	329	4.2
PROPANE	19	0.2
PROPANE LPG	404	5.1
ISO BUTANE	334	4.2
NORMAL BUTANE	393	5.0
LIGHT NAPHTHA RAFFINATE	492	6.3
HEAVY NAPHTHA RAFFINATE	122	1.5
SULFUR	355	4.4
AMMONIA	82	1.0
BENZENE	144	1.7
TOLUENE	385	4.8
XYLENE	285	4.2
CARBON BLACK OIL	1,891	24.1
ELECTRODE BINDER	123	1.6
CALCINED COKE	1,870	23.8
PHENOLS	124	1.6
AMINES	12	0.2
TOTAL	7,840	100.0

FIGURE 15 - PETROCHEMICAL FEEDSTOCKS FROM A SOLVENT REFINED COAL

FEEDSTOCK	1985		1990		1995	
	MM LB/YR	PERCENT OF 1974 CONSUMPTION	MM LB/YR	PERCENT OF 1974 CONSUMPTION	MM LB/YR	PERCENT OF 1974 CONSUMPTION
ETHANE	2,400	15	6,500	40	13,700	90
PROPANE + LPG	3,100	20	8,300	55	17,600	115
NAPHTHA (RAFFINATE)	4,500	80	12,100	220	25,500	470
BENZENE	1,000	9	2,800	25	6,000	50
TOLUENE	2,800	40	7,600	110	16,000	235
XYLENE	2,100	45	5,600	125	11,900	265

FIGURE 16 - EXAMPLES OF POTENTIAL FOR PRODUCTION OF PETROCHEMICAL FEEDSTOCKS FROM COAL

COLLECTED WORK NO. 25

PETROCHEMICAL FEEDSTOCKS FROM COAL

J. B. O'Hara, E. D. Becker, N. E. Jentz & T. Harding
The Ralph M. Parsons Company
Pasadena, California

From CHEMICAL ENGINEERING PROGRESS - June 1977

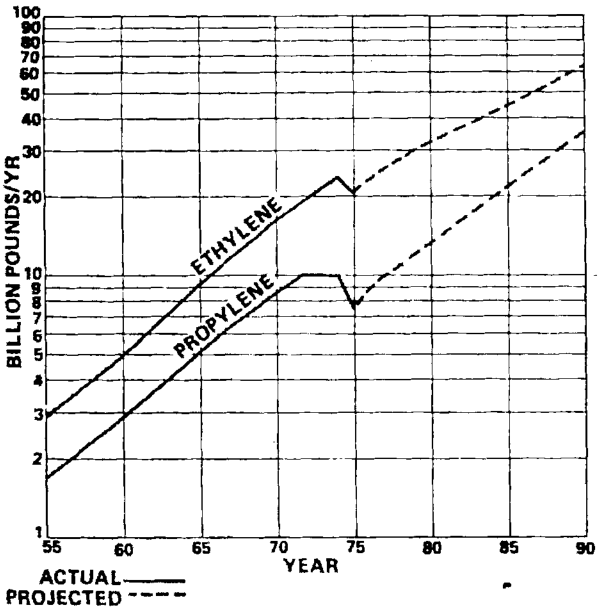


Figure 1. U.S. production of ethylene and propylene.

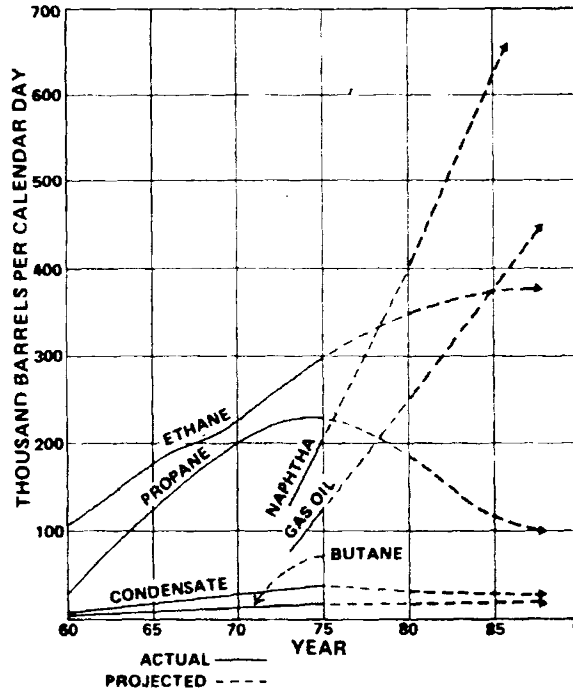


Figure 2. Feedstock consumption for ethylene production, U.S. supply.

COAL PROCESSING:

Petrochemical Feedstocks From Coal

Here are some ways that coal conversion technology can become a major feedstock supplier, a development that could alter the structure of the petrochemical industry.

J. B. O'Hara, E. D. Becker, N. E. Jentz, and T. Harding
The Ralph M. Parsons Co., Pasadena, Calif.

During the last few years it has become virtually impossible to speak about the outlook of any segment of our economy without using the word "shortage." The petrochemical industry is no exception. Specifically, new sources of feedstocks to supply the olefins plants of the future must be acquired. Current alternatives include possibilities for recovering light hydrocarbons from domestic natural gas, domestic crude oil products, imported crude oil, imported refined products, and imported liquefied natural gas. Each potential source presents problems when future feedstock requirements and uncertainties of supply and price are taken into account. Therefore, at least one more source should be considered: coal.

This article describes the potential of petrochemical-from-coal complexes that can produce olefin feedstocks, benzene-toluene-xylenes (BTX), petrochemicals, and chemicals.

First, let us examine why the search for feedstock supplies has become more difficult in recent years. One of the reasons is the average growth rate of petrochemicals during the last 10 years, which has been above the average of the U.S. economy. Current projections indicate that this growth will continue in the immediate future. (*1*) Another factor is the 1973 embargo with its resulting four-fold increase in world energy prices and resulting cause for continuing concern over reliability of supply. And still another cause is the decrease in production rates of U.S. natural gas and crude oil.

These factors, and others, have raised serious questions about keeping light-hydrocarbon-fed Gulf Coast olefin plants on-stream, and about feedstock supplies for plants yet to be built. However, incentives exist to continue to explore the alternatives.

Through the early 1970's, U.S. ethylene-based petrochemical plants depended largely on light hydrocarbons, including ethane, propane, and to a lesser extent butane, for their feedstocks. The results of the 1973 oil embargo and other developments have forced a revision in plans for the mix of future feedstocks.

Ethane will still find a major outlet as an ethylene feedstock. It must, however, adjust to a number of factors including decreased natural gas supply, "drier" natural gas production with lower ethane concentrations from the deeper wells, and partial compensation by a trend to deeper extraction of ethane by cryogenic means to increase the percentage recovery of available ethane.

For example, the percent ethane extraction by absorption had been in the range of 25%, while cryogenic extraction can raise this to 85% recoveries. (2) Greater impact has appeared in the propane and butane feedstocks because of the large increase in energy prices, and the continued expansion in the importance of liquefied petroleum gas in residential and agricultural markets.

Lacking assurance of an increase in the supply of light feedstocks, projections now show a significant shift to naphtha and gas oil in coming years. As an example, one source has projected that naphtha and gas oil will supply approximately 55% of the ethylene feedstock in 1985 as contrasted to about 13 percent % in 1970. (*1*) Others project greater naphtha/gas oil contribution. Probable cost of naphtha and gas oil by 1980 is in the range of $16 to $18/bbl. (3) and ethylene prices in the range of 20¢ to 25¢/lb. are possible. (4)

The result is a significant incentive to provide an assured, competitively priced source of light feedstocks to again permit construction of new olefin plants using these feeds. Incentives include fixed capital investment savings, simplicity of product mix resulting in reduced marketing complications, and potential attractive net production economics.

Coal conversion technology can produce these light hydrocarbons. It can also produce naphtha and gas oils suit-

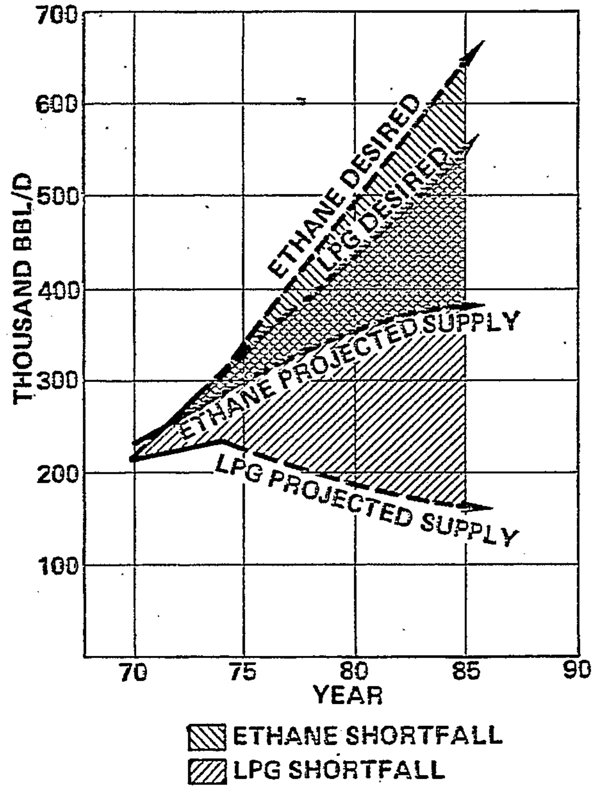


Figure 3. Light hydrocarbon ethylene feedstocks vs. projected supply.

Table 1. Projected product yields, oil/gas plant, SRC II conversion technology.

Product	wt. %	bbl. (or mm std. cu. ft.)/ 100 tons Coal	Million B.t.u./100 tons Coal
SNG	11.0	(0.47)	490
NH_3	0.3	—	—
Sulfur	3.6	—	—
LPG	2.6	28.4	112
Naphtha	3.6	26.3	151
Fuel Oil	31.7	158.0	1,097
Ash	11.8	—	—
Total			1,850
Dissolver H_2 Consumption (wt. % MF)	4.7		
Liquid/SNG Ratio— B.t.u./B.t.u.	2.8		

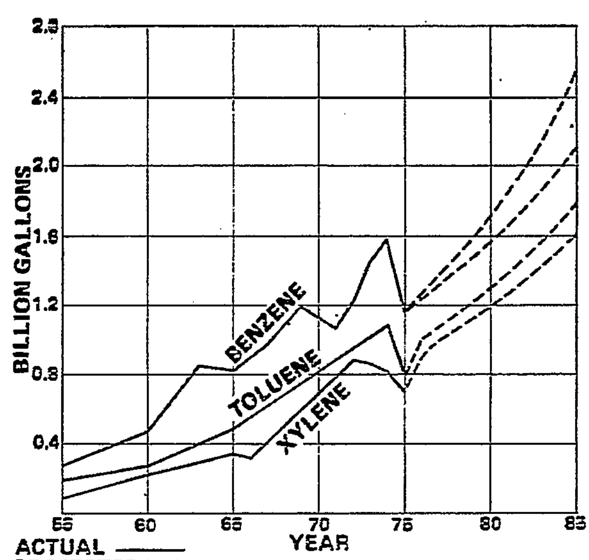


Figure 4. U.S. production of benzene, toluene, and xylene.

Table 2. Projected product yields, SRC II conversion technology.

	Liquid/SNG Ratio, B.t.u. Basis					
	≈2.8			≈5.9		
Product	wt. %	bbl. (or mm std. cu. ft.)/ 100 tons Coal	Million B.t.u./ 100 tons Coal	wt. %	bbl. (or mm std. cu. ft.)/ 100 tons Coal	Million B.t.u./ 100 tons Coal
SNG	11.0	(0.47)	400	5.7	(0.24)	250
NH_3	0.3	—	—	0.1	—	—
Sulfur	3.6	—	—	3.5	—	—
LPG	2.6	28.4	112	0.5	5.7	20
Naphtha	3.6	26.3	151	3.9	34.0	155
Fuel Oil	31.7	158.0	1,097	39.9	205.6	1,288
Ash	11.8	—	—	11.8	—	—
Totals			1,850			1,713

Table 4. Oxygenates stream analysis.

Component	Vol. %
Acetone	3.2
Methyl Ethyl Ketone	0.7
Methanol	6.0
Ethanol	67.6
Propanol	18.0
Butanol	2.4
Amyl Alcohol	1.1
Higher Alcohols and Other Oxygenates	1.0
Total	100.0
Specific Gravity	0.8
HHV (dry), B.t.u./lb.	13,100

Table 3. Projected product yields sprayed catalyst Fischer-Tropsch Plant.

Product	wt. %	bbl. (or mm std. cu. ft.)/ 100 tons Coal	Million B.t.u./100 tons Coal
SNG	22.3	(0.875)	909
Sulfur	3.4	—	—
LPG	1.2	12.0	49
Naphtha	8.0	68.3	333
Distillate Fuel	9.5	71.2	385
Oxygenates	1.9	13.1	39
Ash	7.8	—	—
Total			1,715

able for olefin feedstock use and BTX's. Definition of effective procedures to convert coal to these products, as well as the economics, could result from the major program now underway in the U.S. to develop viable coal conversion technology. This article describes several ways that this conversion can be done and comments on the market impact.

Feedstock demand

The historical data for production of ethylene and propylene, as well as projections to 1990, are shown in Figure 1. (*5, 6, 7*) The growth rate in ethylene demand for the period 1955-65 was approximately 11%; in 1965-75 about 7.5%; and projections are that it will be approximately 7% over the period 1975 to 1985. (*2*) Current information indicates that of the nine ethylene plants to be completed in the U.S. in the late 1970s, only one, or at most two, will use light hydrocarbon feedstocks, with the remainder depending primarily on naphtha and gas oil. (*8*)

The trend lines for consumption of ethane, propane, butane, condensate, naphtha, and gas oil as feedstocks for ethylene production for the period of 1960-75 is illustrated in Figure 2. Also shown are projections for the period of 1975-90 (*1, 9, 10*), which are based on the intent to supply the feedstocks from domestic natural gas and crude oil sources, recognizing that the feedstocks will be priced on the basis of energy values set in the international and domestic markets.

Figure 3 (*1, 2, 11, 12*) shows an apparent incremental market demand for light hydrocarbon feedstocks if an assured competitive supply were available. It assumes that the olefin plant operators had the same supply options that existed in the pre-embargo early 1970s and that future olefin plants would use the same feedstock percentages that existed in 1970. Based on the number of olefin plants planned to 1985, the desired amount is shown by the upper curve. The lower curve represents projections of domestic light feedstocks expected to be available for the olefin manufacture. The difference is a shortfall of supply that potentially could be satisfied by an alternative source such as coal conversion. This shortfall provides a target.

Historical and projected data for production of the BTX's are shown in Figure 4. (*5, 13, 14*) In the following discussion, emphasis will be placed on the ethylene feedstocks and BTX's because these materials represent more than 50% of the total tonnage of petrochemicals. Further, these materials can be effectively produced by coal conversion.

The information presented above indicates there is a market for additional ethylene feedstock supply at a competitive price; production of BTX at a competitive price also provides incentives. The next step, then, is to find ways that these materials might be produced using indigenous coal as a raw material.

Incentives to use coal as a base include the fact that it is our largest indigenous fossil fuel reserve. Potentially, this resource could supply industry expansion needs without necessity to depend on imports. Historically, the chemical industry grew up based on coal as a raw material. Coal was rapidly phased out when more convenient and lower cost natural gas and crude oil became available.

The situation is now different; we cannot be assured of control over adequate supplies of oil and gas, the costs for these sources have increased markedly, and there is uncertainty regarding future supply and cost. It is, therefore, opportune to survey the results that are available now and can be expected in the future from the U.S. coal conversion development program to define how these technologies can be applied to produce petrochemicals, petrochemical feedstocks, and chemicals, in addition to fuels. As in the case of oil, future petrochemicals-from-coal complexes must be intimately intertwined with the energy supply/demand and product value picture. The petrochemical and the energy industries must be viewed as interdependent entities.

The following discussion is based on the premise that incremental future assured competitive supply of light hydrocarbon feedstocks will have a value. They would permit the petrochemical industry to build and operate simpler, less capital intensive olefin plants, resulting in a less complex marketing scope because fewer coproducts would be produced than are now planned for naphtha and gas oil feeds. (*6*) In a sense, it would permit the industry to continue its growth pattern based on feedstock decisions structured in pre-embargo days. The technical ability to use our ample indigenous coal resources to provide these feedstocks therefore deserves attention. Careful economic assessment will quickly follow acceptance of the existence of technical capability to supply a significant part of the feedstock market.

The incentive also exists to develop viable procedures to produce liquid petrochemical feedstocks in the naphtha and gas oil range, but for slightly different reasons. They include:

- Greater independence from imported crude oil supplies.
- Potential assured high-quality feedstock supply.
- A broad product/feedstock interaction between the coal conversion and the petrochemical industries.

All of the feedstocks can be produced by coal conversion techniques. Several possibilities to achieve this will be described and two specific plant configurations complete with projected product quantities will be presented.

Process variations

Inputs for development of the process configurations to be discussed come from many sources including data from the ERDA-sponsored pilot plants and smaller experimental units plus four conceptual commercial coal conversion plant designs that Parsons has completed under ERDA-sponsorship. These include:

1. A 1973 "Clean Boiler Fuels" design that converted 10,000 ton/day of coal to approximately 25,000 bbl./day of a product mix consisting of naphtha, distillate fuel, and heavy fuel oil. (*15*) This used SRC II hydroliquefaction technology.
2. A 1974 "Oil and Power by COED-Based Technology" design that converted about 25,000 ton/day of coal to approximately 30,000 bbl./day of distillate fuel plus about 850 mW of electrical energy. (*16*) This used low pressure fluid bed coal pyrolysis and steam-oxygen char gasification.
3. A 1976 "Oil/Gas" mixed product (liquid/gas) design that will convert about 36,000 ton/day of coal to SNG, LPG, naphtha, and fuel oil. (*17*) This used high conversion SRC II hydroliquefaction technology.
4. A 1976 "Fischer-Tropsch" mixed product (liquid/gas) design that will convert about 30,000 ton/day of coal to SNG, LPG, naphtha, diesel fuel, oxygenates and fuel oil. (*18*) This used an "indirect" synthesis technique: full gasification of the feed coal to produce synthesis gas, purification of the syngas, and synthesis of liquids using Fischer-Tropsch technology.

A key point: The liquid products from the first three designs mentioned above tend to be rich in aromatics while Fischer-Tropsch products tend to be rich in aliphatics. The two processes could, therefore, complement each other in a complex to be designed to produce both olefin feedstocks and BTXs.

Liquefaction technology is preferred for large volume production of olefin feedstocks and BTX from coal. A 1975 paper (*19*) listed typical yields from uncatalyzed hydroliquefaction (SRC), catalyzed hydroliquefaction (H-Coal), pyrolysis (COED), extraction (CSF) and Fischer-Tropsch (SYNTHOL fluid bed conversion) processes. This type of yield data remains as important inputs to a feedstock production analysis effort.

Two additional types of product yield structure will now be added to this inventory based on the results of recent work. Table 1 lists the plant yields predicted for a high

Table 5. Fischer-Tropsch naphtha characteristics.

Characteristic	Light Naphtha	Heavy Naphtha
Gravity, °API	85.5	71.3
Distillation ASTM, °F:		
IBP	96	186
10	115	208
50	137	236
90	159	266
EP	185	300
Color, Saybolt	+30	+30
Viscosity at −30°F, CS	0.8	1.7
Aniline Point	140°F	160°F
Sulfur, wt. %	nil	nil
Mercaptan Sulfur	nil	nil
Doctor Test	neg	neg
Corrosion, Copper Strip		
At 150°F	1	1
At 210°F	1	1
Oxygen Content, wt. %	0.2 max	0.6 max
H_2S, wt. %	nil	nil
Alcohol and Oxygenates, %	<0.1	<0.1
Octane Number		
F-1 Clear	45.6	—
F-2 Clear	40.6	—
Reid Vapor Pressure, lb./sq. in. abs.	10	—
IP Smoke, mm	over 30	over 30 (IP designation)
Yield Nitrogen, parts/million	nil	nil
UOP "K"	12.85	12.65

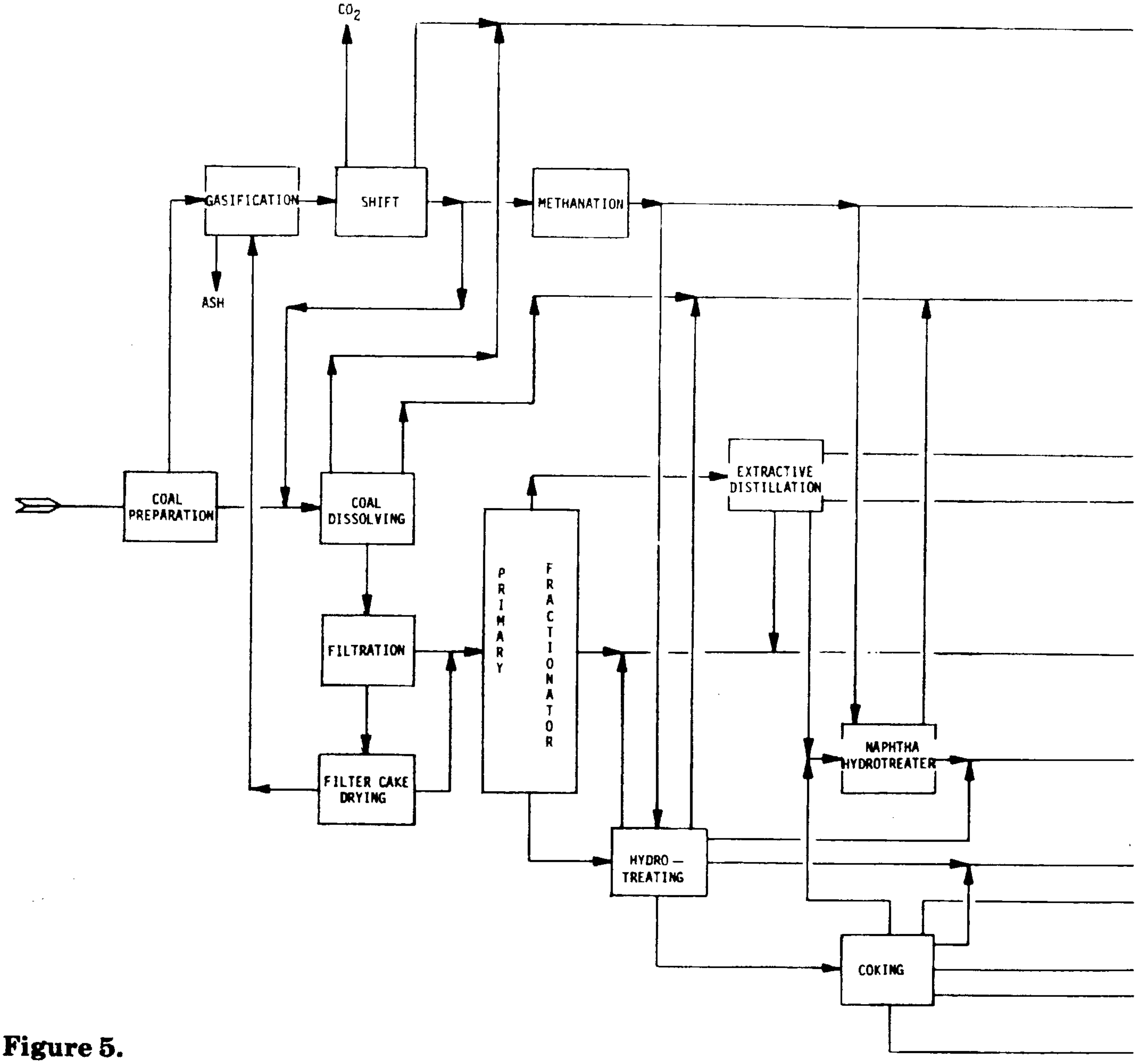
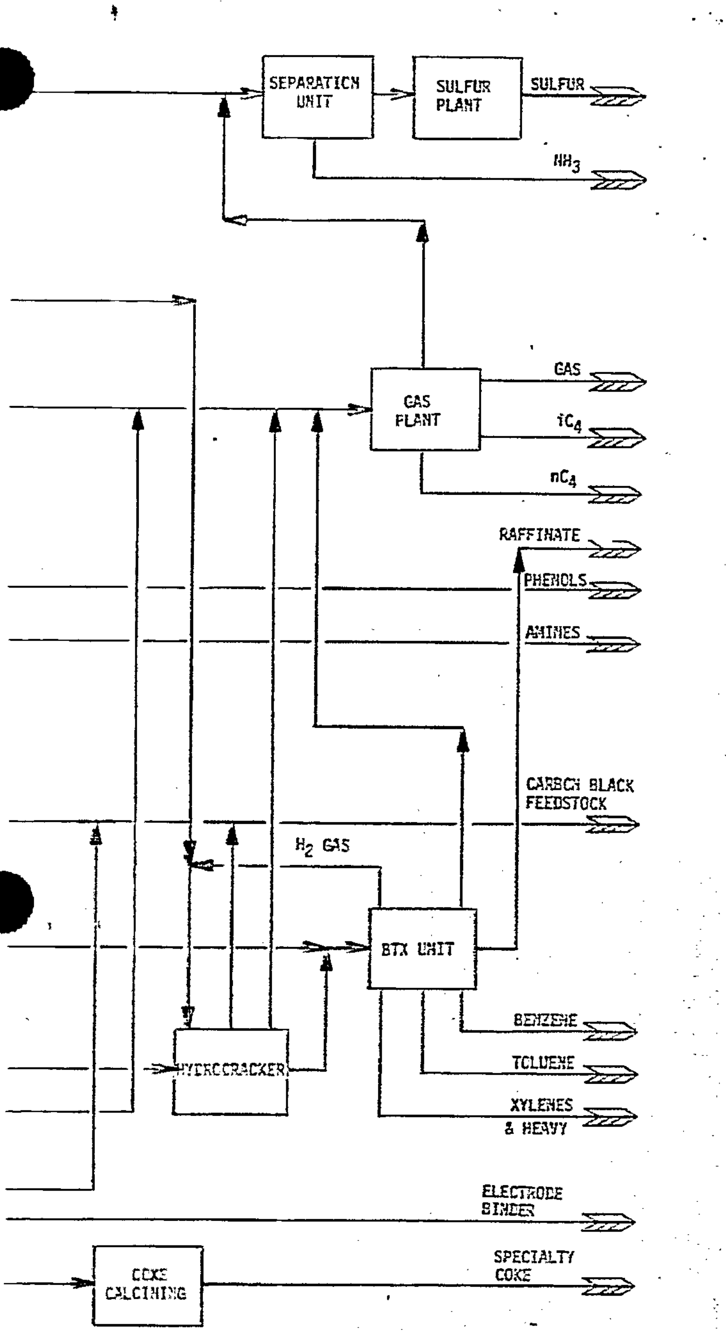


Figure 5.
A possible process sequence for petrochemical feedstock production.

hydrogen consumption pseudocatalytic SRC II mode of operation designed to produce a product slate consisting of a liquid/SNG product ratio of approximately 2:8 on a B.t.u. basis. (*17*) The SRC II conversion technology is under development by the Pittsburg and Midway Coal Mining Co., a division of Gulf Oil Corp., under ERDA sponsorship. Table 1 shows SNG yield to be about 11 wt. %, 400°F end point naphtha to be about 4%, and total liquids to be about 38% based on dry coal feed. The coal dissolver hydrogen consumption was projected to be 4.7 wt. % of moisture-free coal.

The potential flexibility of this technology in petrochemical feedstock production applications is illustrated by Table 2, which compares the Table 1 yield structure with an alternate case with significantly less light hydrocarbon production. (*20*).

Projected yields of a Fischer-Tropsch plant using flame-sprayed iron catalyst are listed in Table 3. (*18*) These yields were projected based on experimental work done on the conversion process at ERDA's Pittsburgh Energy Research Center. (*21*) They were developed as a part of a conceptual commercial design/economic evaluation of a U.S. version of a Fischer-Tropsch plant that Parsons developed under ERDA sponsorship.



To illustrate, Table 3 shows that the potential yields of SNG, LPG, naphtha, and distillate fuel from Fischer-Tropsch are predicted to be about 22.3, 1.2, 8.0, and 9.5 wt.-%, respectively, based on dry Illinois No. 6 coal feed. For reference, the projected composition of the oxygenate stream, a potential source of chemicals, is shown in Table 4 and the projected naphtha characteristics are presented in Table 5.

Coal based-plant concepts

The next step is a look at several coal-based plant concepts that could supply petrochemical feedstocks in quantity.

An earlier presentation on the subject of petrochemical feedstocks from coal described an SRC II-mode based complex that produced approximately 19 wt. % of C_2–C_4s and about 8% naphtha suitable for ethylene feedstock use. (*19*) BTX yield was approximately 11%. For reference, the projected flow-scheme and plant yield structures are shown in Figure 5 and Table 6, respectively. These feedstocks could be produced in sufficient quantity to have a market impact.

A more comprehensive combined energy/petrochemicals production complex has recently been defined to achieve economy of scale plus efficiency of operations. This concept is the counterpart of an oil-based petrochemical refinery. Its implications include large fixed capital investments and broad fuels/petrochemicals marketing requirements in return for the promise of improved production economics.

A process sequence with these characteristics is shown in Figure 6. It incorporates the following major process steps:

1. Coal liquefaction by SRC II mode of conversion.
2. Coal liquefaction by Fischer-Tropsch conversion.
3. Coal/coal residue gasification to produce syngas and hydrogen.
4. Hydrocracking of coal liquids.
5. Naphtha desulfurization.
6. Naphtha reforming.
7. Aromatics extraction.
8. Hydrodealkylation.
9. Ethylene production.
10. Fischer-Tropsch liquid hydrogenation.
11. Methanation.

Table 6. Petrochemical feedstocks from a solvent refined coal complex.

Component	Production Rate, ton/day (Basis: 10,000 ton/day Process Coal Feed)	Product Composition, wt. %
Methane	476	6.1
Ethane	329	4.2
Propane	19	0.2
Propane LPG	404	5.1
Isobutane	334	4.2
Normal Butane	393	5.0
Light Naphtha Raffinate	492	6.3
Heavy Naphtha Raffinate	122	1.5
Sulfur	355	4.4
Ammonia	82	1.0
Benzene	144	1.7
Toluene	385	4.8
Xylene	285	4.2
Carbon Black Oil	1,891	24.1
Electrode Binder	123	1.6
Calcined Coke	1,870	23.8
Phenols	124	1.6
Amines	12	0.2
Totals	7,840	100.0

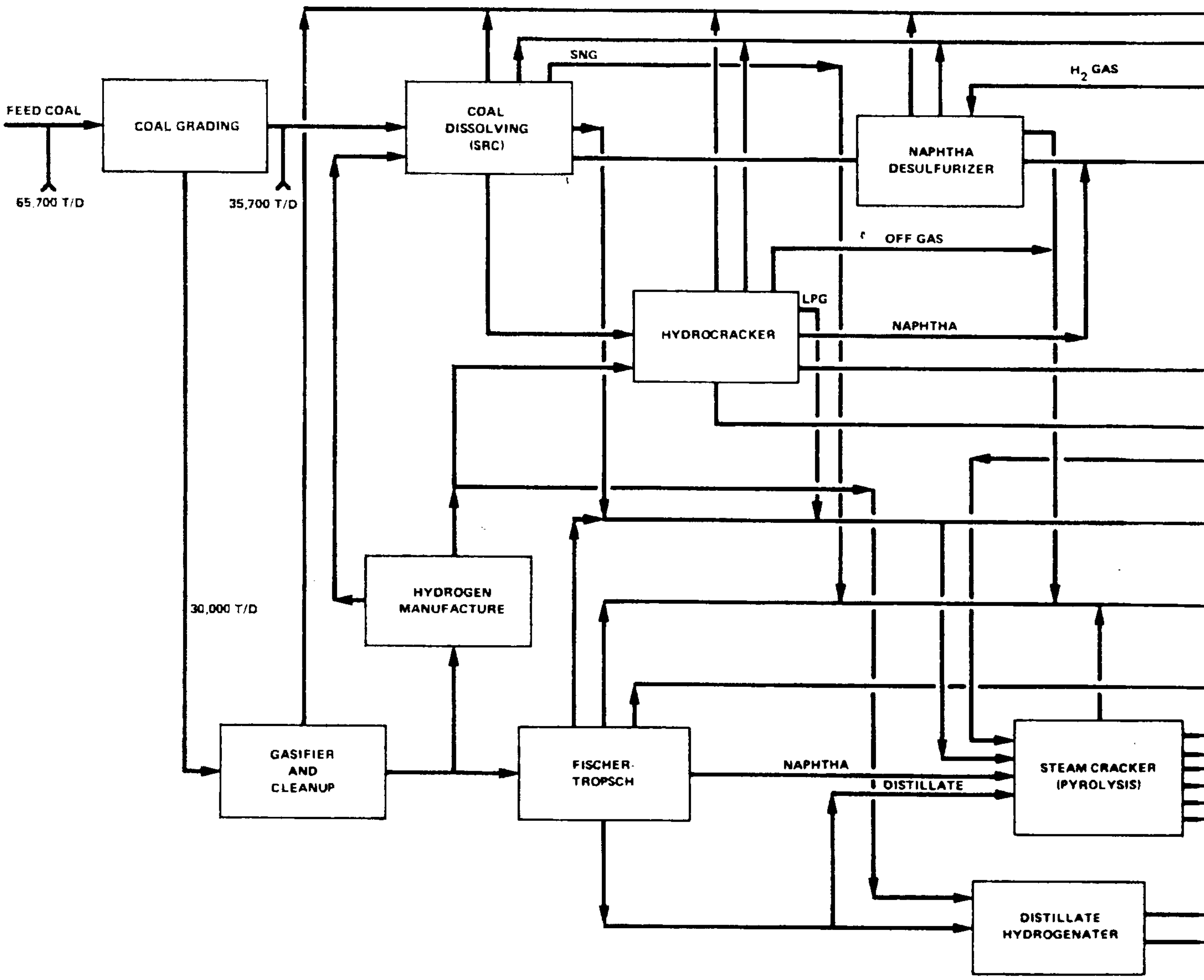


Figure 6. Block flow diagram of a . . .

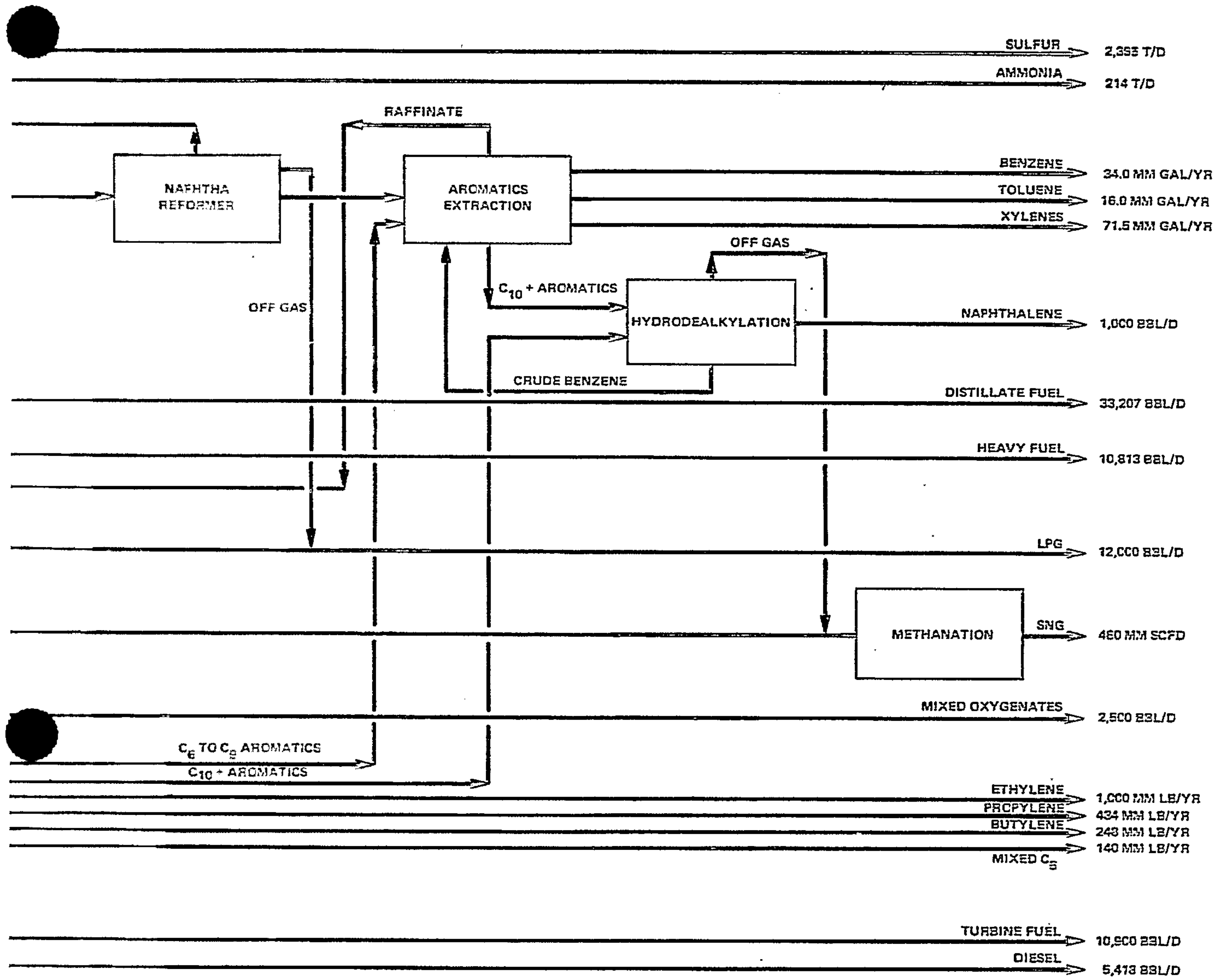
The projected material balance based on an Illinois No. 6 seam coal feed rate of 66,000 ton/day is shown in Table 7.

The number of such petrochemical/energy complexes required to produce 10% of the projected ethylene requirements in the period 1980–1990 is illustrated in Table 8, which indicates that 3.2 of these complexes would be required in 1980 and 6.4 in 1990. Projected market penetrations of propylene and benzene for 1990 are shown to each be in the range of 8%. In addition, a number of other fuel and chemical products are produced.

Principal points regarding the flow schemes in Figures 5 and 6 include the promised flexibility of the coal conversion technology when mated with supporting hydrocarbon processing/petrochemical production techniques—and that the complexes are energy self-sufficient. Inputs are coal, air and water with all utilities internally generated. Obviously, there are a number of alternate process combinations that may be used, depending on the objectives of the complex. The inclusion of the ethylene plant provides the advantages of in-plant processing of pyrolysis gasoline to increase the BTX yield and in that sense simplifies marketing activities.

In conclusion

There are incentives to develop additional assured economically competitive domestic supplies of petrochemical feedstocks, including the light hydrocarbons that historically have been the preferred feeds for U.S. olefin plants and BTX's. Based on current supply/demand pro-



... coal-to-petrochemicals plant.

jections, heavier feedstocks in the naphtha-gas oil range will dominate future olefin production with required higher capital investment and more complex product mixes and broader marketing requirements. The introduction of coal conversion as an important feedstock supplier could affect the future structure of the petrochemical industry. This article suggests ways in which coal conversion plus supporting process steps can be used to supply petrochemical feedstocks and petrochemicals.

During hydroliquefaction coal conversions, light hydrocarbons and aromatic-rich liquids are produced and can be processed to yield desirable feedstocks. Fischer-Tropsch conversion can also be made to produce light hydrocarbons, and in this case, heavier liquids that are aliphatic in nature. Projected yield structures have been presented, and a conceptual complex is described which uses these products to produce ethylene, BTX and a number of fuel and chemical products. Six-to-seven of these complexes, each feeding approximately 65,000 ton/day of coal, could supply 10% of the projected 1990 U.S. ethylene requirements. Companion benzene production would be approximately 8% of the projected U.S. market requirements.

Results of preliminary analysis work to date provide a base for more detailed technical and economic analysis of this potentially important extension of the results of the current national priority ERDA-sponsored fuels-from-coal development program. Until other assured, economical sources of the preferred feedstocks can be found, this alternative should continue to be carefully considered.

Table 7. Production quantities for a coal-to-petrochemicals plant.

Component	Production Rate, ton/day, (Basis: 66,000 ton/day Process Coal Feed)		Product Composition, wt. %
SNG	460	million std. cu. ft./day	38.52
Sulfur	2,375	ton/day	8.38
Ammonia	214	ton/day	0.75
Benzene	34	million gal./yr.	1.34
Toluene	16	million gal./yr.	0.62
Xylenes	71.5	million gal./yr.	2.79
Naphthalene	1,000	bbl./day	0.71
Distillate Fuel	33,207	bbl./day	16.13
Heavy Fuel	10,813	bbl./day	7.73
LPG	12,000	bbl./day	4.04
Mixed Oxygenates	2,500	bbl./day	1.22
Ethylene	1,000	million lb./yr.	5.34
Propylene	434	million lb./yr.	2.32
Butylenes	248	million lb./yr.	1.33
C_6	140	million lb./yr.	0.75
Turbine Fuel	10,900	bbl./day	5.53
Diesel	5,418	bbl./day	2.50
Total			100.00

Table 8. Projected market penetration, olefins and BTX from coal (Basis: ethylene production = 10% U.S. demand).

	1980		1990	
Product	Production Rate	% Market	Production Rate	% Market
Ethylene, million lb./yr.	3,200	10.0	6,400	10.0
Propylene, million lb./yr.	1,400	8.2	2,800	8.2
Benzene, million gal./yr.	110	6.4	220	7.5
Toluene, million gal./yr.	50	3.8	100	4.6
Xylene, million gal./yr.	230	19.2	460	23.0

Acknowledgment

We would like to thank the Major Facilities Project Management Div. of ERDA-Fossil Fuel Energy, and in particular Messrs. N. P. Cochran and D. Garrett, for their support and guidance. Also, the development of the conceptual process described here represents the works of a number of people. Credit is particularly due to Messrs. A. Bela, S. M. Fass, G. H. Hervey, and D. G. Reynolds of The Ralph M. Parsons Co. #

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O'Hara


Becker


Jentz


Harding

SECTION 4
EMERGING ENERGY TECHNOLOGY COMPARISONS

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