LOW-NO$_x$ COMBUSTORS FOR ALTERNATE FUELS CONTAINING SIGNIFICANT QUANTITIES OF FUEL-BOUND NITROGEN

W. D. Clark
D. W. Pershing
G. C. England
M. P. Heap

Energy and Environmental Research Corporation
8001 Irvine Boulevard
Santa Ana, California 92705

ABSTRACT

This paper summarizes data generated on two EPA-sponsored programs concerned with the development of low-NO$_x$ combustors for high nitrogen containing fuels. EPA Contract 68-02-3125 is concerned with NO$_x$ production and control from liquid fuels containing significant quantities of bound nitrogen. It was found that fuel nitrogen content is the primary composition variable affecting fuel NO formation and that emissions from both petroleum and alternative liquid fuels correlate with total fuel nitrogen content. Conditions were identified which allow high-nitrogen fuels to be burned satisfactorily with minimal NO$_x$ emissions. Certain coal-derived fuel gases may contain ammonia. Data is presented from a series of bench-scale reactors designed to minimize the conversion of this ammonia to NO$_x$. Lowest NO$_x$ emissions were produced in a rich/lean combustor utilizing either a diffusion flame or a catalyst in the fuel-rich primary stage.

SECTION 1

INTRODUCTION

Combustion of liquid fuels derived from petroleum sources accounts for a significant fraction of fossil fuel consumption in stationary combustors. As petroleum reserves grow smaller, the United States is projected to place heavy reliance on coal, the most abundant fossil fuel available, in the search for new energy supplies. Coal can be burned directly or converted into either a liquid or a gaseous fuel. The potential for low sulfur emissions makes combustion of gasified coal an environmentally attractive alternative to direct-fired coal combustion. However, low-Btu coal gases can contain ammonia concentrations as high as 0.38 percent (1). In a conventional combustor, much of this ammonia may be converted to nitrogen oxides resulting in significant pollutant emission: up to 1370 ng/J (3.2 lbm/10^6 Btu) for full conversion of NH_3 to NO_2.

A balanced fuel economy necessitates that in the future many industrial users will burn petroleum and coal- or shale-derived liquid fuels. Since these liquid fuels have relatively high nitrogen content and low hydrogen-to-carbon ratios, there will be the potential for adverse environmental impact due to the increased emission of combustion-generated pollutants unless preventative measures are taken (1-2). The pollutant of major concern in this paper is nitrogen oxides (NO_x). The paper addresses the impact of switching from conventional fuels to alternative gaseous or liquid fuels and of the mechanisms of combustion modification techniques used to control NO_x emissions.

Alternative liquid fuels can be broadly classified as those synthesized from the products of coal gasification, and those derived directly as liquids. The fuels in the first category tend to be clean, low-boiling-point fuels such as alcohols, and are essentially free from nitrogen and sulfur; thus, their impact upon pollutant emissions is minimal. The liquids in the second category may be compared to crude petroleum oils containing a wide range of hydrocarbon compounds with boiling points from 300°K to greater than 900°K. The bound nitrogen content of crude synfuels is generally higher than petroleum crudes, and for many applications it might be necessary to upgrade the fuel by removing the nitrogen. Recognizing that alternative liquid fuels

contain more bound nitrogen than the petroleum fuels that they would be
replacing, one key factor in their production is to what extent combustion
modification will allow control of NO_x emissions and reduce the necessity
for substantial denitrification, thereby reducing the cost of synfuels.

Nitrogen oxides produced during combustion emanate from two sources.
Thermal NO is formed by the fixation of molecular nitrogen and its forma-
tion rate is strongly dependent upon temperature (3). Fuel NO is formed by
the oxidation of chemically-bound nitrogen in the fuel by reactions with a
weak temperature dependence, but a strong dependence upon oxygen avail-
ability (4-5-6-7). Thus, those emission control techniques which minimize
peak flame temperature by the addition of inert diluents (e.g., cooled recycled
combustion products or water addition) minimize thermal NO formation, but
have a minor impact upon fuel NO production. Staged heat release (staged com-
bustion) provides the most effective NO_x control technique for nitrogen-
containing fuels because fuel NO formation is mainly dependent upon local
stoichiometry. It can be accomplished either by separating the combustion
chamber into two zones and dividing the total combustion air into two streams,
or by appropriate burner design which promotes localized fuel-rich conditions.

Minimizing fuel NO_x formation requires the existence of a fuel-rich
primary combustion zone to maximize the conversion of fuel nitrogen to
molecular nitrogen since the fate of fuel-bound nitrogen is strongly con-
trolled by the reactant stoichiometry. Many studies (8-12) have shown that
under fuel-rich conditions the efficiency of conversion to N_2 increases
significantly. Thus, there are two fuel nitrogen reaction paths leading to
the production of N_2 or NO, namely:

Path A.   Fuel-lean

$$XN + Oxidant \rightarrow NO + ...$$

Path B.   Fuel-rich

$$XN + ... \rightarrow N_2 + ...$$
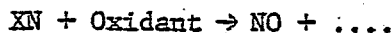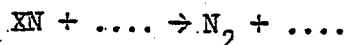
The objective of staged combustion emission control techniques is the pro-
vision of conditions which maximize N_2 production via Path B. Two factors of
practical importance are the residence time and the stoichiometry required to
maximize N_2 production in the fuel-rich primary zone.

If the residence time is insufficient, then the original fuel nitrogen species will exist in the gaseous state as some XN compound which can be converted to NO in the second-stage heat release zone. The stoichiometry required to achieve minimum XN concentrations at the exit of the primary stage will be determined by (1) the rate of evolution of nitrogen species from the fuel; (2) the inevitable distribution of stoichiometries from fuel-rich to fuel-lean which occurs because the primary zone is supplied by a diffusion flame; and (3) the overall temperature of the primary zone. From equilibrium considerations the total fixed nitrogen (TFN given by $NO + HCN + NH_3$) is a minimum at approximately 65 percent theoretical air with levels less than 10 ppm depending upon temperature and fuel C/H ratio. Exhaust NO_x emissions are considerably greater than levels predicted by equilibrium, suggesting the existence of kinetic limitations in the fuel-rich primary stage.

NO_x formation during combustion of alternate fuels is not well-understood; however, recent test results have indicated that replacing a petroleum oil with a coal- or shale-derived liquid may result in a major increase in NO_x emissions. Bench-scale experiments (13) have shown that the smoke and combustion characteristics of the SRC-II coal liquids are equivalent to light oil, but uncontrolled NO_x emissions are high due to the 0.8 to 1.2 percent N in the fuel. Pilot-scale SRC-II studies (14-16) have demonstrated that both fuel blending and staged combustion are effective in reducing NO_x emissions and that improved atomization, increased preheat, and increased excess O_2 increase NO_x. Full-scale testing (17) has confirmed the need for optimized combustion modifications. Similar results have also been achieved during bench-scale (18) and field tests (19) with shale-derived liquids.

SECTION 2

EXPERIMENTAL SYSTEMS

The experimental systems used to investigate NO_x formation from gaseous and liquid fuels have been described in detail elsewhere and only a brief summary will be presented in this paper.

LBG GAS STUDIES

The apparatus for the bench-scale experiments can be divided into four subsystems: LBG supply, modular combustors, sample train and control systems. A simplified schematic of the facility can be seen in Figure 1.

Synthetic LBG was produced from hot air premixed with vaporized water and heptane passed through a catalytic reformer. The reformer was operated at pressures between 6.4 and 11.9 atmospheres at a stoichiometry of 45 percent theoretical air, the richest stoichiometry attainable without excessive sooting. The water acts as a diluent to maintain the maximum catalyst bed temperature at around $1370^{\circ}K$. The reformer product gas passed through a variable heat exchanger, cooling it to the desired preheat temperature. Ammonia and methane are added to trim the gas to the desired fuel nitrogen and hydrocarbon content. The LBG passed through a soot filter and into a valve system, controlling the fraction of the LBG which goes to the combustors and the fraction which is bypassed. If none of the gas was bypassed, maximum combustor capacity was 60,000 J/s (200,000 Btu/hr).

The combustors consisted of a series of modules with 5 cm (2 in) ID reaction/flow chambers enclosed in 15 cm (6 in) OD low-density insulation and housed in flanged steel pipe. Primary ignition modules include the catalyst and the diffusion flame. Secondary burnout was achieved in the jet-stirred secondary air injector. Plug flow modules of various lengths allowed control of primary and secondary residence times. The primary ignition modules are shown in Figure 2. In the catalyst module, premixed LBG and primary air passed through a stainless steel flow straightener/flame arrestor and into the graded cell catalyst. The catalyst, supplied by Acurex, consisted of three zirconia honeycomb monoliths of decreasing cell size, coated with nickel oxide. Platinum had been added to the coating of the upstream monolith to promote ignition. In the concentric diffusion flame module, LBG is introduced
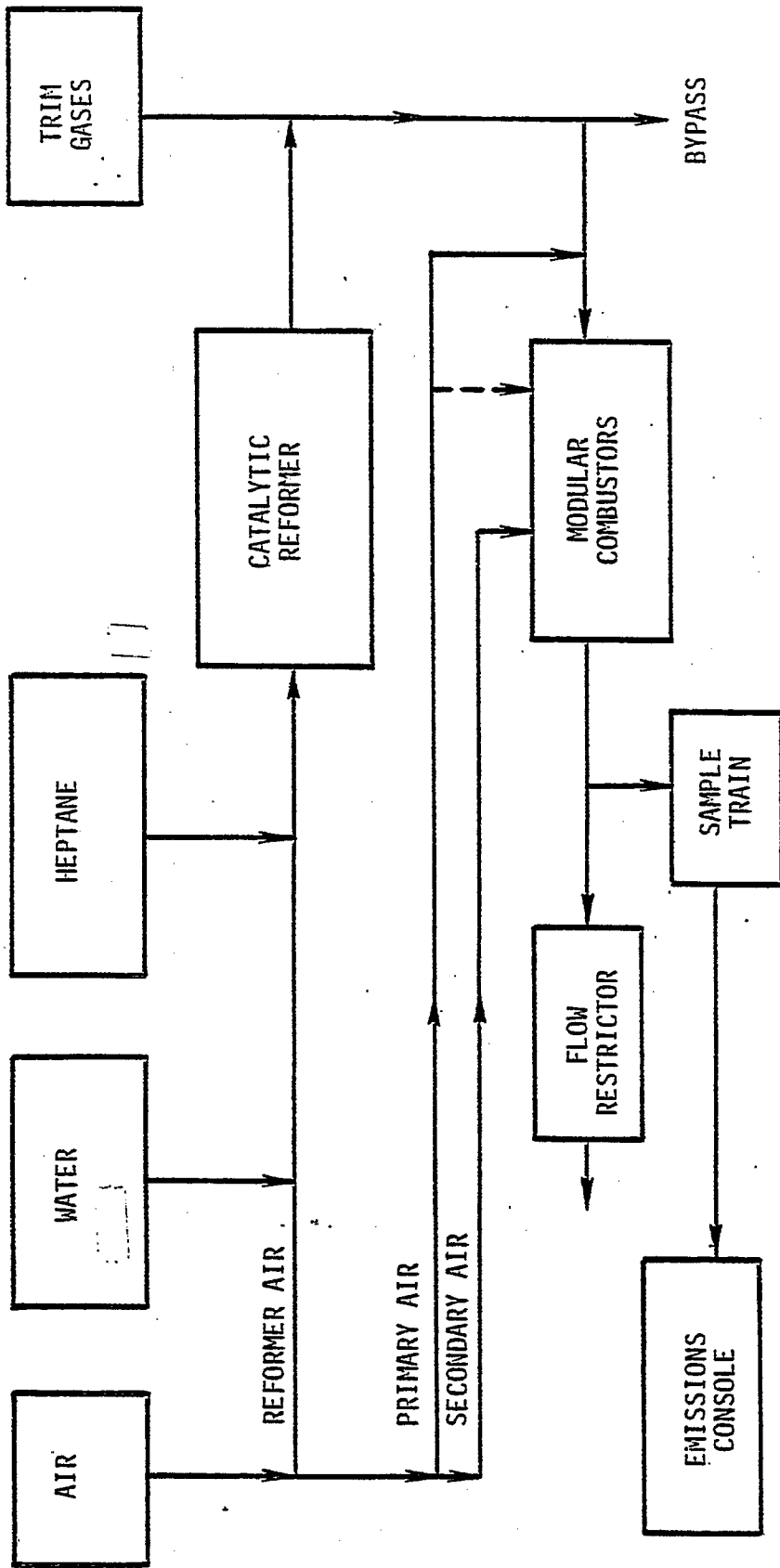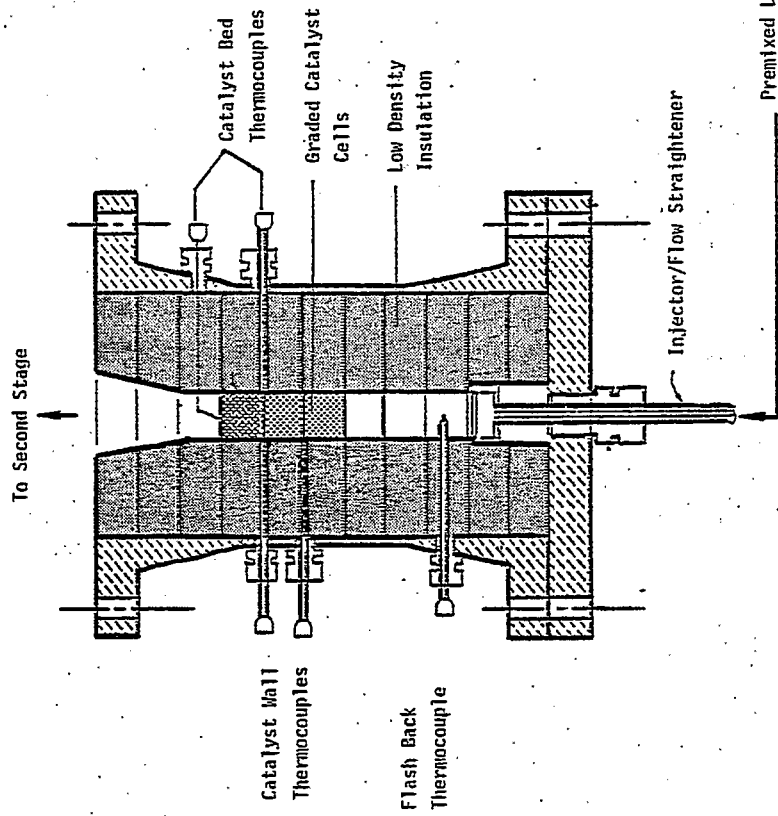
Figure 1. Bench-Scale Pressurized Test Facility.

BENCH-SCALE DIFFUSION FLAME

Plug Flow Module

Low Density Insulation

To Secondary

Primary Air

LBG

Primary Air

BENCH-SCALE CATALYST

Catalyst Bed Thermocouples

Graded Catalyst Cells

Low Density Insulation

To Second Stage

Injector/Flow Straightener

Premixed LBG + Primary Air

Catalyst Wall Thermocouples
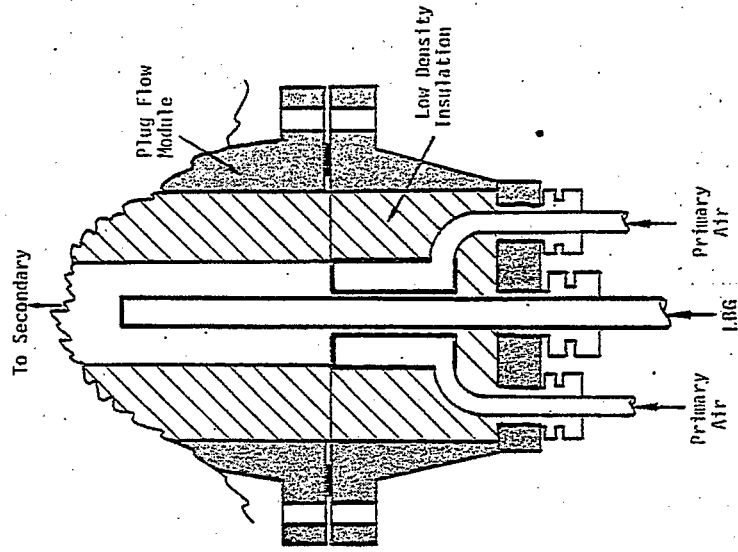
Flash Back Thermocouple

Figure 2. Primary Ignition Modules.

165

through a removable fuel tube of variable diameter. Straightened primary air passes annularly around the fuel tube in the direction of the fuel flow.

Samples are taken in the secondary region, through a water-cooled stainless steel probe situated on the centerline of the flow chamber. The cooling water is preheated and the stainless steel sample lines are wrapped with heat tape to maintain the sample system above the dewpoint of the exhaust gases. The sample stream is throttled to nearly atmospheric pressure.
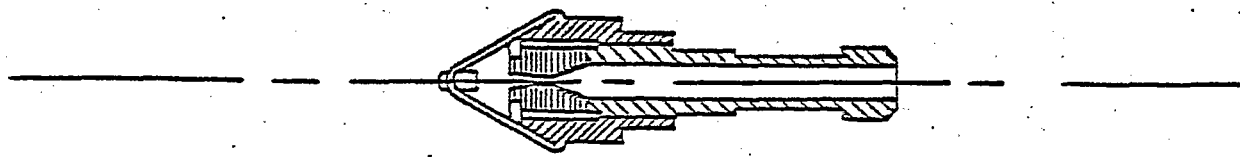
LIQUID-FIRED TUNNEL FURNACE

The downfired tunnel furnace illustrated in Figure 3 was designed to allow utilization of commercially-available spray nozzles, and yet be capable of testing with artificial atmospheres. This combustor, which has been described in detail elsewhere (6), was 2.1 m long and 20 cm in inside diameter. The walls consisted of insulating and high-temperature castable refractories and the full-load firing rate was 0.53 cc/sec, which corresponds to a nominal heat release of 20 kW. All airstreams were metered with precision rotameters. The main combustion air was preheated with an electric circulation heater; the atomization air was not preheated. In certain tests the "air" was enriched or replaced with varying amounts of carbon dioxide, argon, and oxygen, all of which were supplied from high-pressure cylinders.

ANALYTICAL SYSTEMS

Exhaust concentrations were monitored continuously using a chemiluminescent analyzer for NO and NO_x, a NDIR analyzer for CO and CO_2, and a paramagnetic analyzer for O_2. The flue gas was withdrawn from the stack through a water-cooled, stainless steel probe using a stainless steel/Teflon sampling pump. Sample conditioning prior to the instrumentation consisted of an ice bath water condenser and glass wool and Teflon fiber filters. All sample lines were 6.3 mm Teflon and all fittings 316 stainless steel.

In-flame temperature measurements were made with a standard suction pyrometer containing a platinum/rhodium thermocouple. In-flame gas samples were withdrawn with a long, stainless steel water-quench probe. HCN and NH_3 were absorbed in a series of wet impingers and concentrations determined using specific ion electrodes. Sulfide ion interference was minimized by the addition of lead carbonate (20). Hydrocarbons were measured using a water-cooled probe, heated sample line, and an FID analyzer.

Ultrasonic Twin-fluid Atomizer

Thermocouple
Connection

Oil Heater
Connection — Atomizing Air

Oil Pressure
Tap

Combustion
Air

Oil Inlet

Viewing Port

Ultrasonic
Nozzle

Burner Detail

Viewing Port   Burner
                    Section

Insulating
Refractory

Insulating
Block   High-Temperature
             Refractory
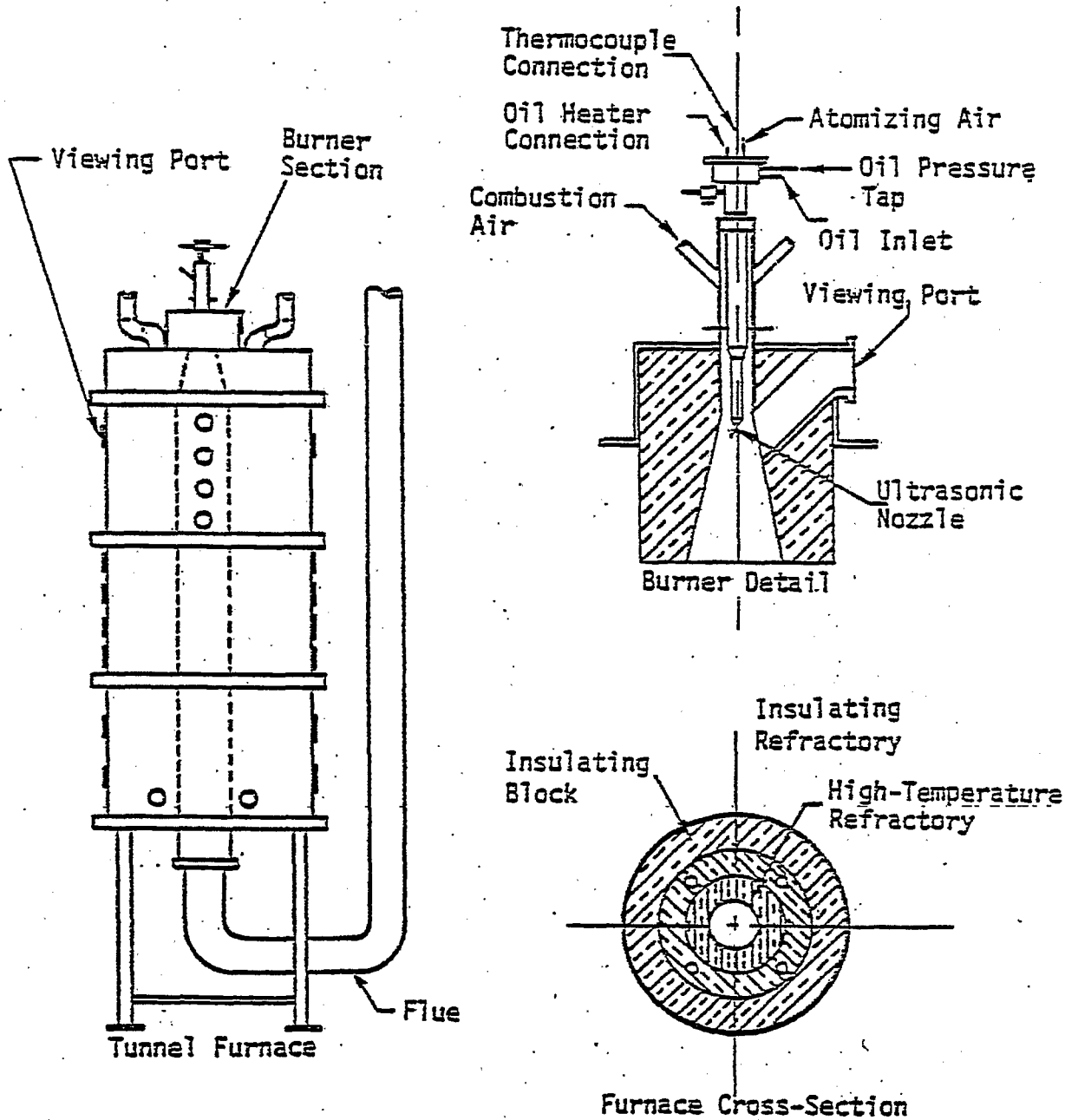
Flue

Tunnel Furnace

Furnace Cross-Section

Figure 3. Details of the Tunnel Furnace System.

LIQUID FUELS

Figure 4 illustrates the wide spectrum of composition for the distillate oils (half-filled symbols), heavy petroleum liquids (open symbols), and alternative liquid fuels (solid symbols) investigated to date. The petroleum-derived fuels had sulfur contents ranging from 0.2 to 2.22 percent with a maximum nitrogen content of 0.86. The nitrogen content of the alternative fuels range from 0.24 to 2.5 percent. Table 1 lists the complete chemical analysis and physical properties of each fuel as determined by an independent laboratory. The shale liquids included crude shale from the Paraho process (A3) and four refined products: diesel fuel marine (DFM, A1) residual fuel oil (A5), a 520-to-850°F distillation cut (A7), and a 5.75/1 medium/heavy SRC-II blend (A6), a heavy SRC-II distillate (A9), and an SRC-II blended with the donor solvent (A4).
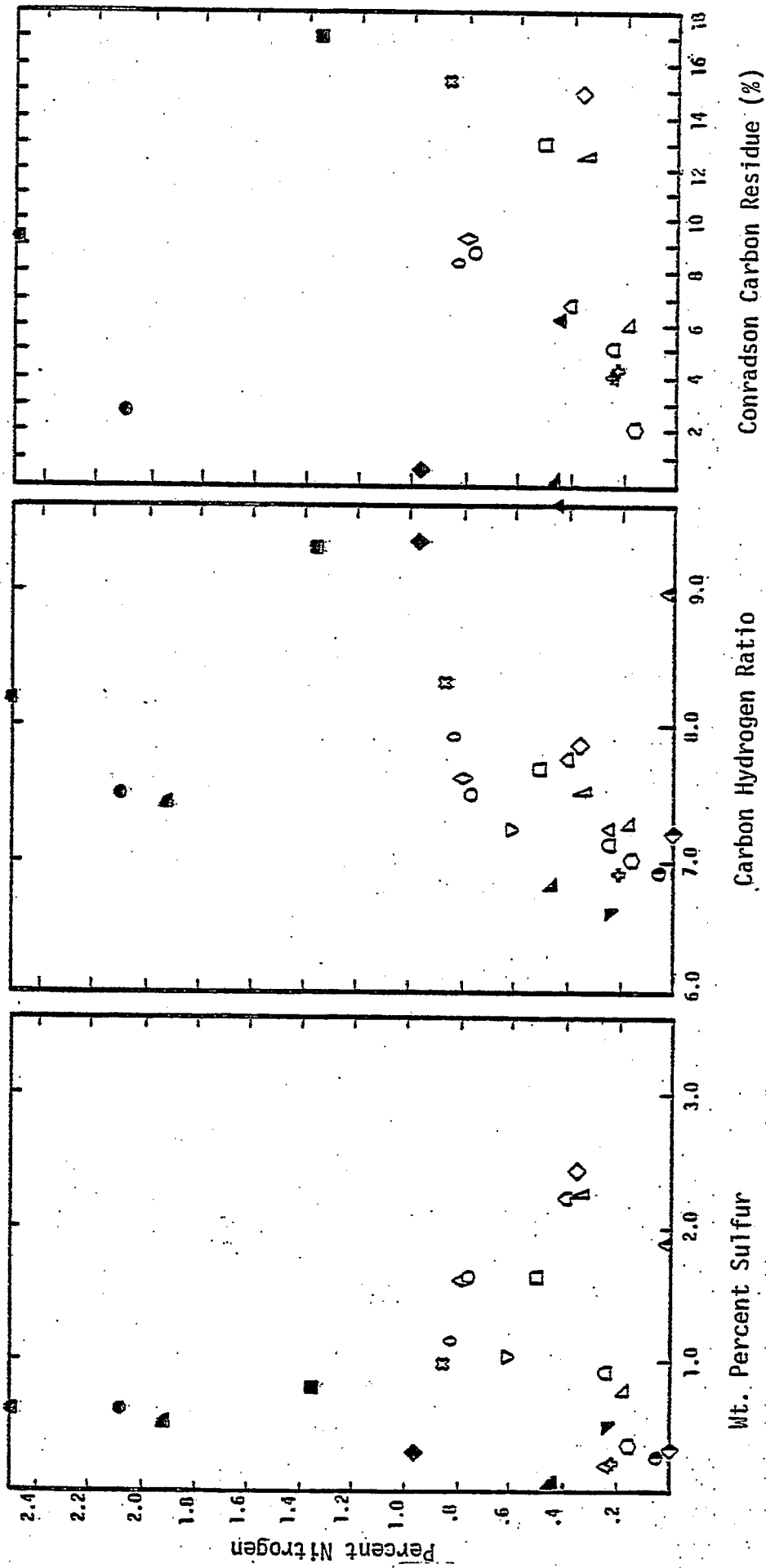
Figure 4. Properties of Fuels Tested.

Table 1. Detailed Fuel Analyses

	D1	D2	D3	R1	R2	R3	R4	R5	R6	R7	R8	R9	R10
	Alaskan Diesel	W. Texas Diesel	California No. 2 Oil	East Coast	Middle East	Low Sulfur No. 6 Oil	Indo/Malaysian	Venezuelan Desulphurized	Pennsylvania (Amarada Hess)	Gulf Coast	Venezuelan	Alaskan	California
Ultimate Analysis:													
Carbon, %	86.99	88.09	86.8	86.54	86.70	86.57	86.53	85.92	84.02	84.62	85.24	86.04	85.75
Hydrogen, %	12.07	9.76	12.52	12.31	11.95	12.52	11.93	12.05	11.21	10.77	10.96	11.18	11.83
Nitrogen, %	0.02	0.026	0.053	0.16	0.18	0.22	0.24	0.24	0.34	0.36	0.40	0.51	0.62
Sulfur, %	0.31	1.88	0.27	0.36	0.67	0.21	0.22	0.93	2.26	2.44	2.22	1.63	1.05
Ash, %	<.001	<.001	<.001	0.023	0.012	0.02	0.036	0.033	0.067	0.027	0.091	0.034	0.030
Oxygen, %	0.62	0.24	0.36	0.61	0.41	0.46	1.04	0.83	1.3	1.78	1.10	0.61	0.71
Conradson Carbon Residue, %				2.1	6.0	4.4	3.98	5.1	12.4	14.8	6.0	12.9	
Asphaltene, %				0.34	3.24	0.94	0.74	2.59	4.04	7.02	8.4	5.6	
Flash Point, °F				205	350	325	210	176	275	155	210	215	
Pour Point, °F				50	40	105	61	40	66	40	50	38	
API Gravity at 60°F	33.1	18.3	32.6	24.9	19.8	25.1	21.8	23.3	15.4	13.2	14.1	15.6	19.5
Viscosity, SSU, at 140°F	33.0	32.0	30.8	131.2	490	222.4	199	113.2	1049	835	742	1,071	246.1
at 210°F	29.5	28.8	29.5	45	131.8	69.6	65	50.5	240	101	196.7	194	70.00
Heat of Combustion:													
Gross Btu/lb			19,330	19,260	19,070	19,110	19,070	18,400	18,520	18,240	18,240	18,470	
Net Btu/lb				18,140	17,980	17,970	17,980	17,300	17,500	17,260	17,400	17,560	
Calcium, ppm				7.1	1.2	9.52	14	8.7	9.2	4.4	9.1	6.9	
Iron, ppm				16	2.6	123.6	16	6.5	13.2	19	11	24	
Manganese, ppm				0.09	0.02	0.46	0.13	0.09	0.10	0.13	0.09	0.06	
Magnesium, ppm				3.7	0.08	2.23	3.6	3.6	3.3	0.4	3.0	1.4	
Nickel, ppm				6.7	13	14.10	19		32.7	29	52	50	
Sodium, ppm				37	0.98	3.74	15		64.5	3.6	32	37	
Vanadium, ppm				14	25	3.11	101		81.5	45	226	67	

170

Table 1. Detailed Fuel Analyses (Continued)

	R11	R12	R13	R14	A1	A2	A3	A4	A5	A6	A7	A8	A9
	California	California	California	California (Kern County)	Shale-Derived DFM	Synthoil	Crude Shale	SRC II Blend	Shale Residual	SRC II	Shale Fraction (520-650°F)	Shale Fraction (+650°F)	SRC II Heavy Distillate
Ultimate Analysis:													
Carbon, %	85.4	85.33	86.66	86.61	86.18	86.30	84.6	89.91	86.71	85.91	85.39	85.92	88.98
Hydrogen, %	11.44	11.23	10.44	10.93	13.00	7.44	11.3	9.27	12.76	8.74	11.53	10.61	7.64
Nitrogen, %	0.77	0.79	0.86	0.83	0.24	1.36	2.08	0.45	0.46	(0.96)	1.92	2.49	1.03
Sulfur, %	1.63	1.60	0.99	1.16	0.51	0.80	0.63	0.065	0.20	0.30	0.72	0.63	0.39
Ash, %	0.043	0.032	0.20	0.030	0.003	1.56	.026	0.004	0.009	0.04	0.002	0.24	0.050
Oxygen, %	0.71	1.02	0.05	0.44	1.07	2.54	1.36	0.30	0.03	4.08	0.44	0.11	1.90
Conradson Carbon Residue, %	0.72	9.22	15.2	8.3	4.1	23.9	2.9	6.18	0.19	0.51	0.07	9.3	
Asphaltene, %	5.18	5.18	0.62	3.92	0.036	16.55	1.33	4.10	0.083	1.73	0.12	4.24	
Flash Point, °F	38	150	100	255	182	210	250	70	235		255	370	265
Pour Point, °F		30	42	65	40	80	80	<-72	90	-55	.70	95	8
API Gravity at 60°F	15.4	15.1	12.6	12.3	33.1	S-1.14	20.3	10.0	29.0	( )	22.3	12.0	1.3
Viscosity, SSU, at 140°F	854	748.0	720	4630	36.1	10,080	97	40.6	54.3	39	62.9	3050	67.2
at 210°F	129	131.6	200	352	30.7	575	44.1	32.6	37.3	( )	41.8	490	41.3
Heat of Combustion:													
Gross Btu/lb	18,470	18,460	18,230	18,430	19,430	16,480	18,290	17,980	19,350	17,100	18,520	18,000	17,120
Net Btu/lb	17,430	17,440	17,280	17,430	18,240	15,800	17,260	17,130	18,190		17,470	17,030	16,240
Calcium, ppm	21	14	90.6	4.4	0.13	1,670	1.5	0.33	4.20		<.05	230	
Iron, ppm	73	53	77.2	15	6.3	109	47.9	3.9	<0.5		2.9	86	
Manganese, ppm	0.8	0.1	0.87	0.15	0.06	6.2	0.17	<0.5	<0.5		0.033	1.3	
Magnesium, ppm	5.1	3.8	31.4	1.1		170	5.40	0.17	0.15		0.021	51	
Nickel, ppm	65	82	88.0	68	0.43	2.6	5.00	<0.5	<0.5		<0.5	7.4	
Sodium, ppm	21	2.6	22.3	3.4	0.09	140	11.71	0.31	2.51		<0.1	11	
Vanadium, ppm	44	53	66.2	39	<.1	6.5	<.3	<1.0	<1.0		<0.2	1.1	

SECTION 3

RESULTS - LBG GAS

Encouragingly low NO_x levels have been achieved on the bench scale utilizing a catalytic reactor and a diffusion flame reactor. An effective fuel nitrogen-reducing catalyst was identified in laboratory-scale experiments and the effects of scale and stoichiometry were examined in the bench-scale experiments. A fuel-lean diffusion flame was identified as an attractive low-NO_x combustor concept in laboratory-scale experiments and effects of scale, stoichiometry, hydrocarbon content of the fuel, fuel tube size, pressure, and primary residence time were examined in the bench-scale experiments.

Effects of catalyst type on fuel nitrogen processing in LBG combustion were examined on the laboratory scale in an unstaged catalytic reactor operated at a constant adiabatic flame temperature of $1473^\circ K$. Figure 5 shows the variable stoichiometry results for two catalysts. The alumina supported platinum catalyst converted almost all fuel nitrogen to NO_x in fuel-lean combustion and had a minimum conversion of 40 percent in fuel-rich combustion. At stoichiometries richer than 60 percent theoretical air, decreasing NO concentrations were overwhelmed by increasing NH_3, and HCN concentrations, causing a sharp rise in ΣXN. The zirconia supported platinum/nickel oxide catalyst converted 80 percent of the fuel nitrogen to NO_x in lean combustion, but had very low conversions in rich combustion. For a 500 ppm NH_3 in LBG dopant level, less than 10 ppm ΣXN were measured at stoichiometries as rich as 40 percent theoretical air. Tests of the platinum/nickel oxide catalyst over a range of adiabatic flame temperatures $(1273-1673^\circ K)$ and with CH_4 as the fuel yielded similar results.

A rich/lean series staged platinum/nickel oxide primary catalytic reactor was selected as a potential low NO_x concept for bench scale testing. The scale-up results were in general agreement with the laboratory-scale results. Figure 6 compares the results of staged combustion of a 500 ppm NH_3 doped LBG at two scales: 1200 and 20,000 J/sec (4000 and 70,000 Btu/hr). Each had high conversions of NH_3 to NO_x in fuel-lean combustion. Minimum conversions occurred
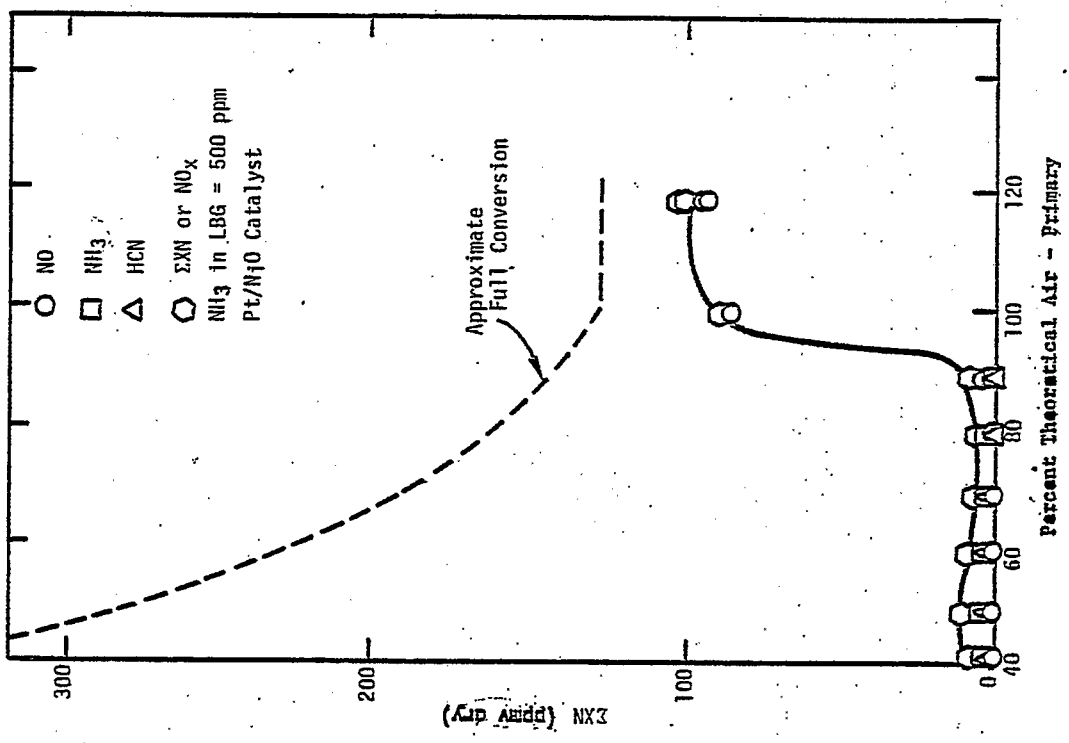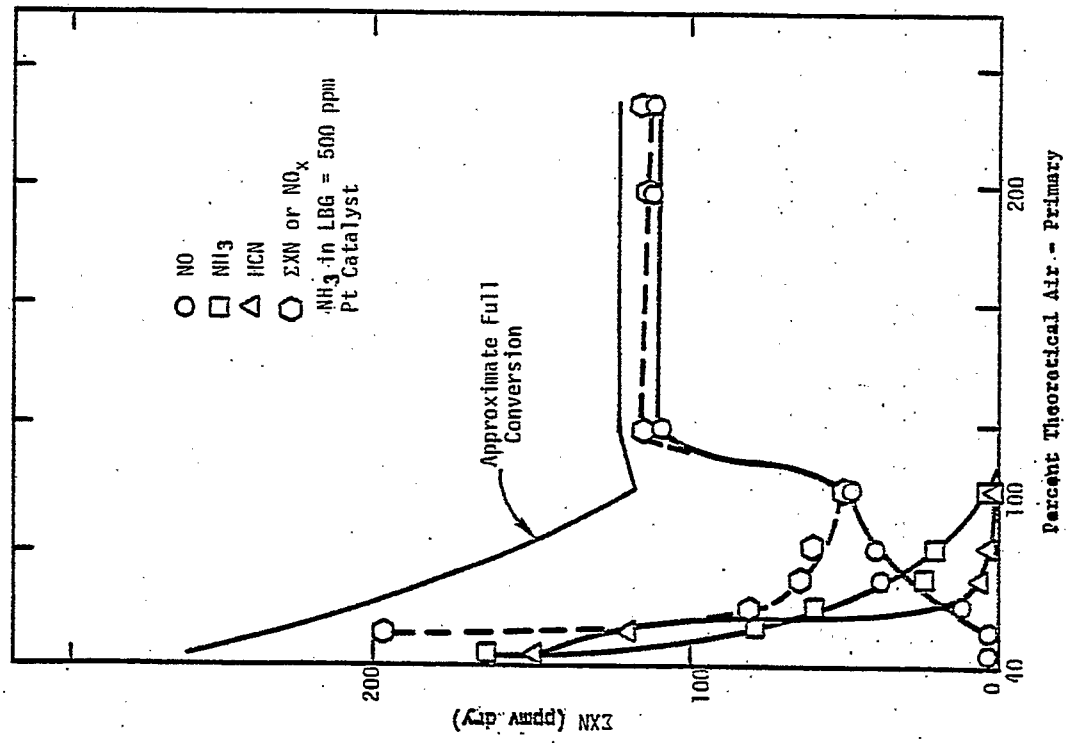
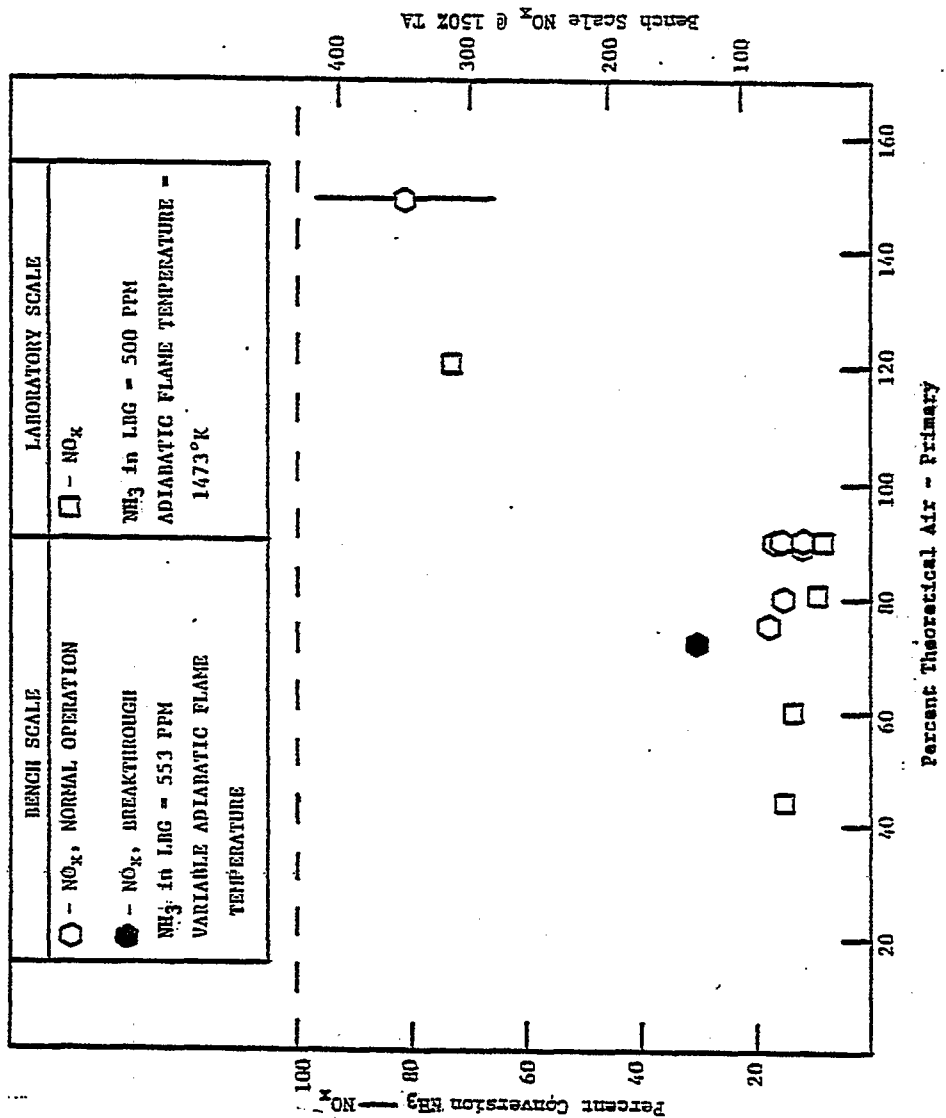172

Figure 5. Laboratory Scale Catalyst Comparison.

Figure 6. Pt/NiO Catalyst Scale-up.

in rich/lean staged combustion when the primary was operated close to stoichiometric. At a primary stoichiometry of 90 percent theoretical air, the laboratory-scale catalytic reactor converted 8 percent of the input NH_3 to NO_x while the bench-scale combustor had an overage conversion of 14 percent. Conversions in the bench-scale combustor remained low (less than 18 percent) over all rich primary stoichiometries under normal operation; but breakthrough occurred if the primary was operated richer than 75 percent theoretical air: the temperatures on the walls of the catalyst monoliths dropped and the conversion rose sharply. Breakthrough was not observed in the laboratory-scale experiment where the adiabatic flame temperature was maintained at a constant $1473^{\circ}K$ by varying the amount of nitrogen diluent in the reactants. The undiluted flame reactor LBG had a higher heating value (HHV) of 6.7×10^6 J/m^3 (180 Btu/ft^3) while the HHV of the bench-scale LBG was only 3.0×10^6 J/m^3 (80 Btu/ft^3). This indicates that raising the heating value of the gas could extend the operating range of the Pt/NiO catalyst, and that catalyst effectiveness is limited by a threshold flame temperature below which breakthrough occurs.

It is difficult to compare the laboratory and the bench-scale diffusion flame combustors. Figure 7 shows laboratory- and bench-scale results for diffusion flame combustion of LBG containing about 500 ppm NH_3 and varying amounts of methane. In the unstaged laboratory-scale experiment, performed at atmospheric pressure in a cold-wall reactor under attached laminar-flow conditions, the hydrocarbon content of the LBG had the most significant effect on XN conversion. Conversions as low as 10 percent were observed for combustion of hydrocarbon-free LBG under nearly stoichiometric conditions. Under richer conditions, conversion increased due primarily to increasing ammonia concentrations. However, under leaner conditions conversions remained quite low. Similar trends were observed in combustion of LBG containing 5 percent methane, but XN conversion was much higher. In the staged bench-scale experiment, performed at 8 atmospheres in a nearly adiabatic combustor under turbulent-flow conditions, effects of hydrocarbon content and stoichiometry were not so pronounced.

The bench-scale flame was not visible and there was no reliable indicator as to whether the flame was attached or lifted. However, throughput and tube size ranged from conditions where the flame should definitely be
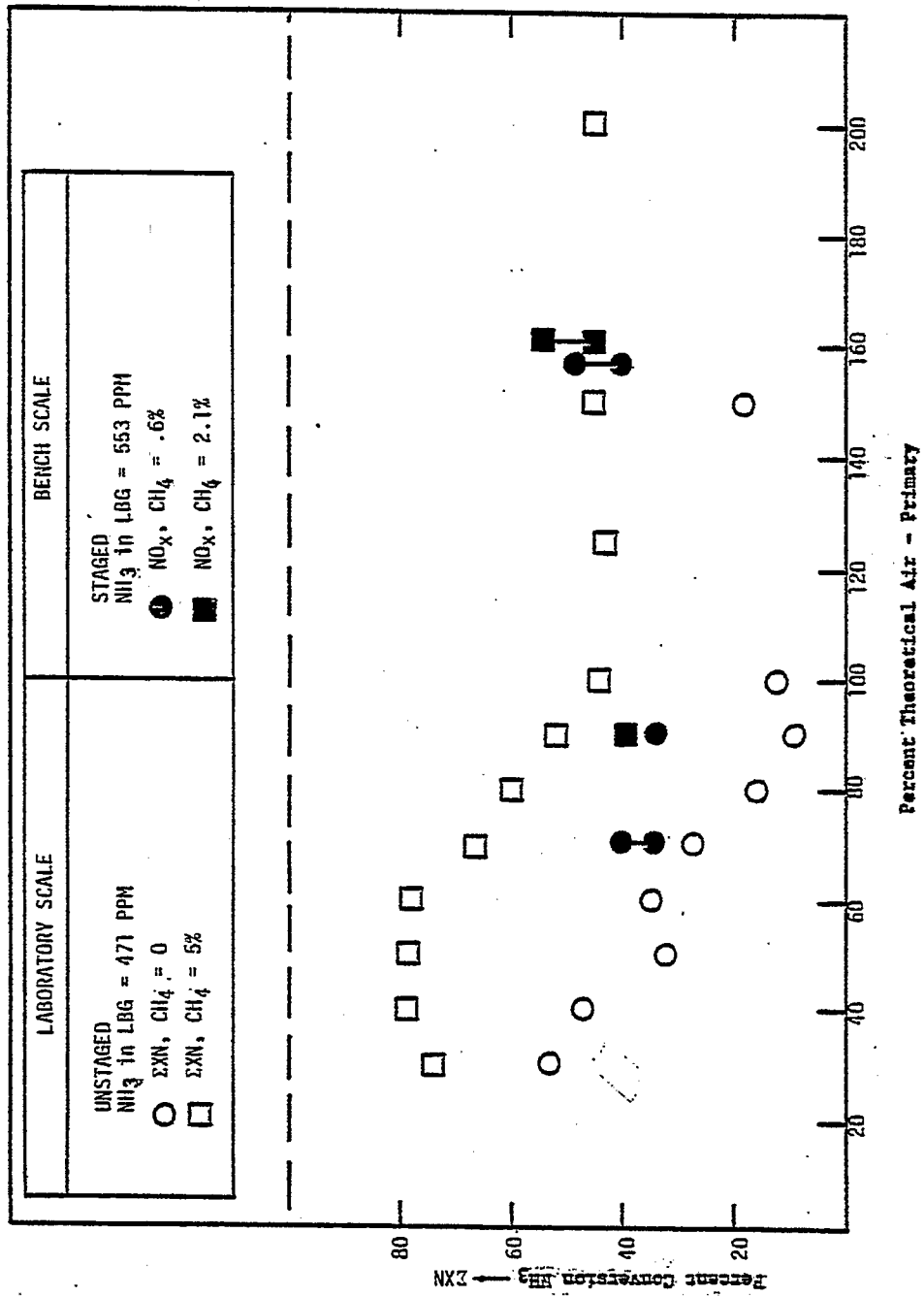
175

Figure 7. Diffusion Flame Scale-up.

attached to conditions where the flame should definitely be lifted. No sharp changes were observed in NO_x emissions or in other measured parameters, indicating that the attached/lifted transition was not an important factor. This agreed with previous variable-throughput laboratory-scale tests of a hydrocarbon-containing diffusion flame, where a smooth NO_x transition was observed as the flame became detached (3).

Figure 8 shows the effect of fuel tube size on XN conversion in the bench-scale diffusion flame operated at 8 atmospheres. In constant-pressure operation at a fixed stoichiometry, fuel flow and primary residence time were independent of fuel tube size, while Reynolds number was inversely proportional to the fuel tube I.D., fuel tube size had little effect on XN conversion in fuel-rich combustion. However, in lean combustion, increasing tube size (decreasing Reynolds number) decreased NH_3 conversion to NO_x. Increased tube size also decreased the NO_x noise level (high frequency concentration fluctuation shown by the error bars in the figure), perhaps an indication of flame stability.

Figure 9 shows the effect of pressure on rich/lean and lean diffusion flames. In the bench-scale system, pressure is maintained by passing the exhaust gases through a critical-flow orifice. For a fixed stoichiometry, fuel flow and Reynolds number are proportional to pressure while primary residence time is independent of pressure. The staged tests were performed at a primary stoichiometry of about 95 percent theoretical air. For low-hydrocarbon LBG, NH_3 conversion to NO_x remained constant at 33 percent over pressures ranging from 4 to 8 atmospheres. For LBG containing 2.1 percent CH_4, conversions remained constant around 40 percent with changing pressure. The lean tests were performed at a stoichiometry of about 150 percent theoretical air. Noise levels were higher than in the staged case. Conversions increased slightly with increasing pressure in low hydrocarbon combustion. Little change in conversion was seen with changing pressure for the 2.1 percent CH_4 LBG.

Primary residence time appeared to have the most pronounced affect on XN conversion in a staged diffusion flame. Residence time was varied at constant pressure by changing the pressure control orifice size. In constant-pressure operation at a fixed stoichiometry, fuel flow and Reynolds number were inversely proportional to primary residence time. Figure 10 shows XN
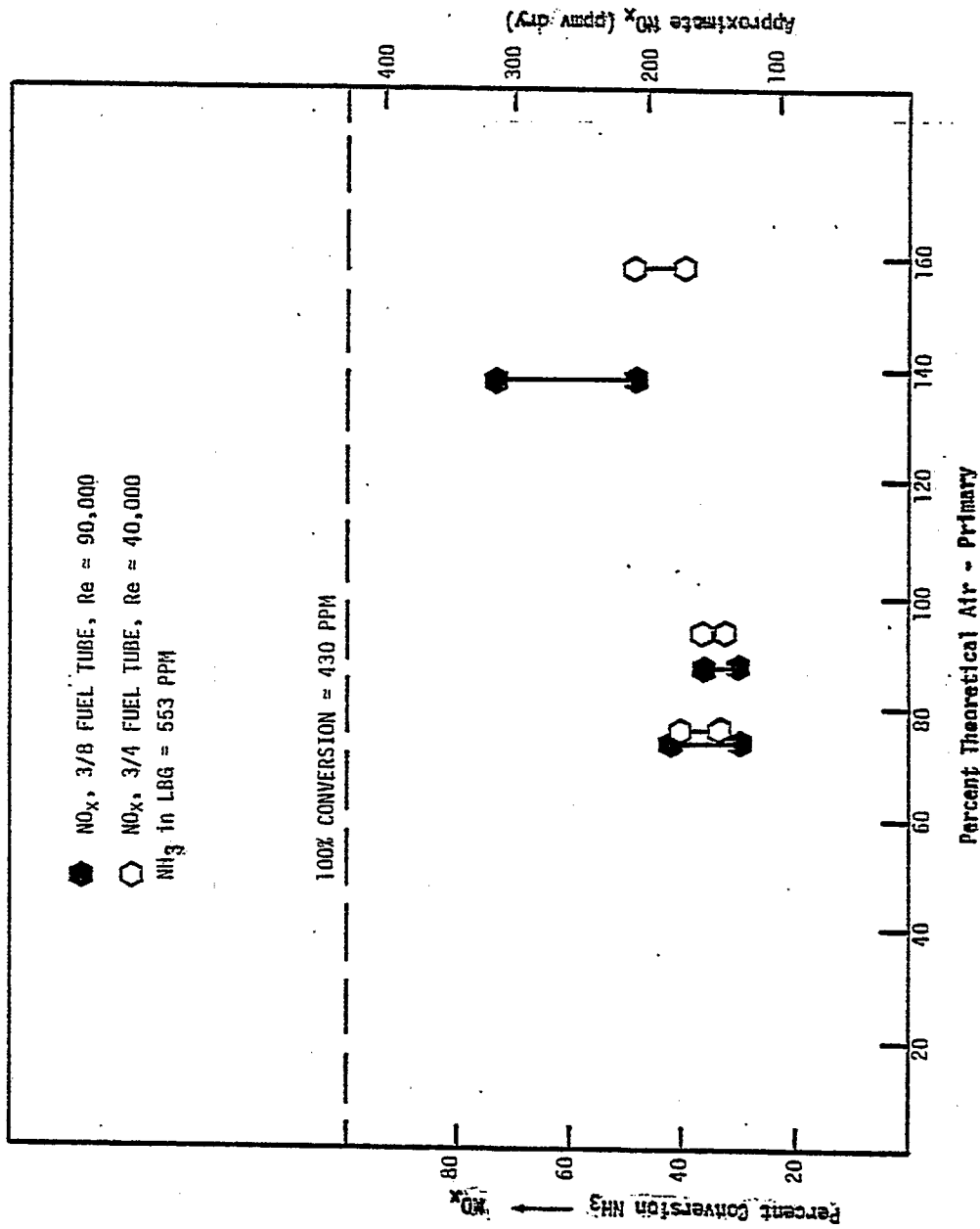
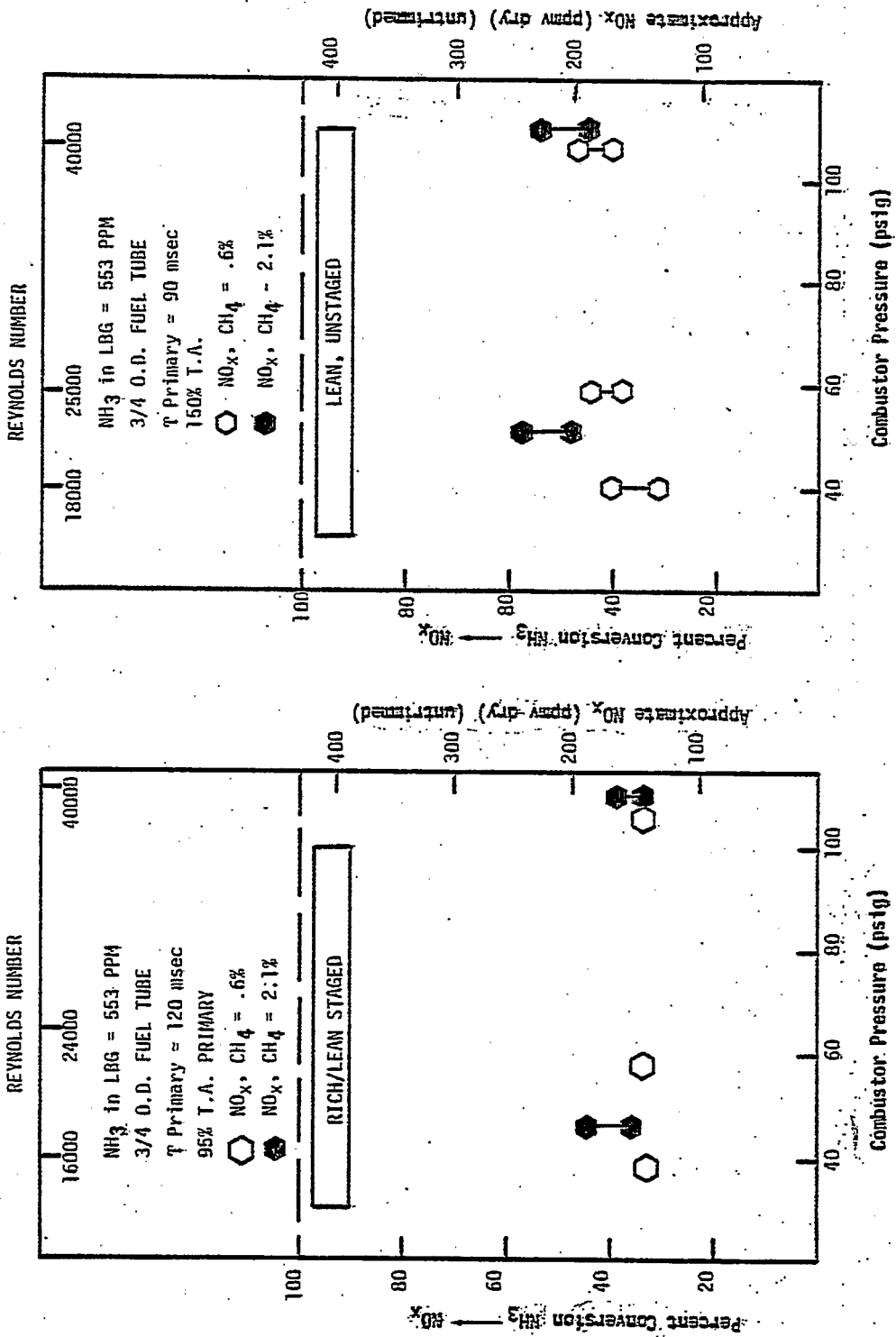Figure 8. Bench-Scale Diffusion Flame: Effect of Reynolds Number.

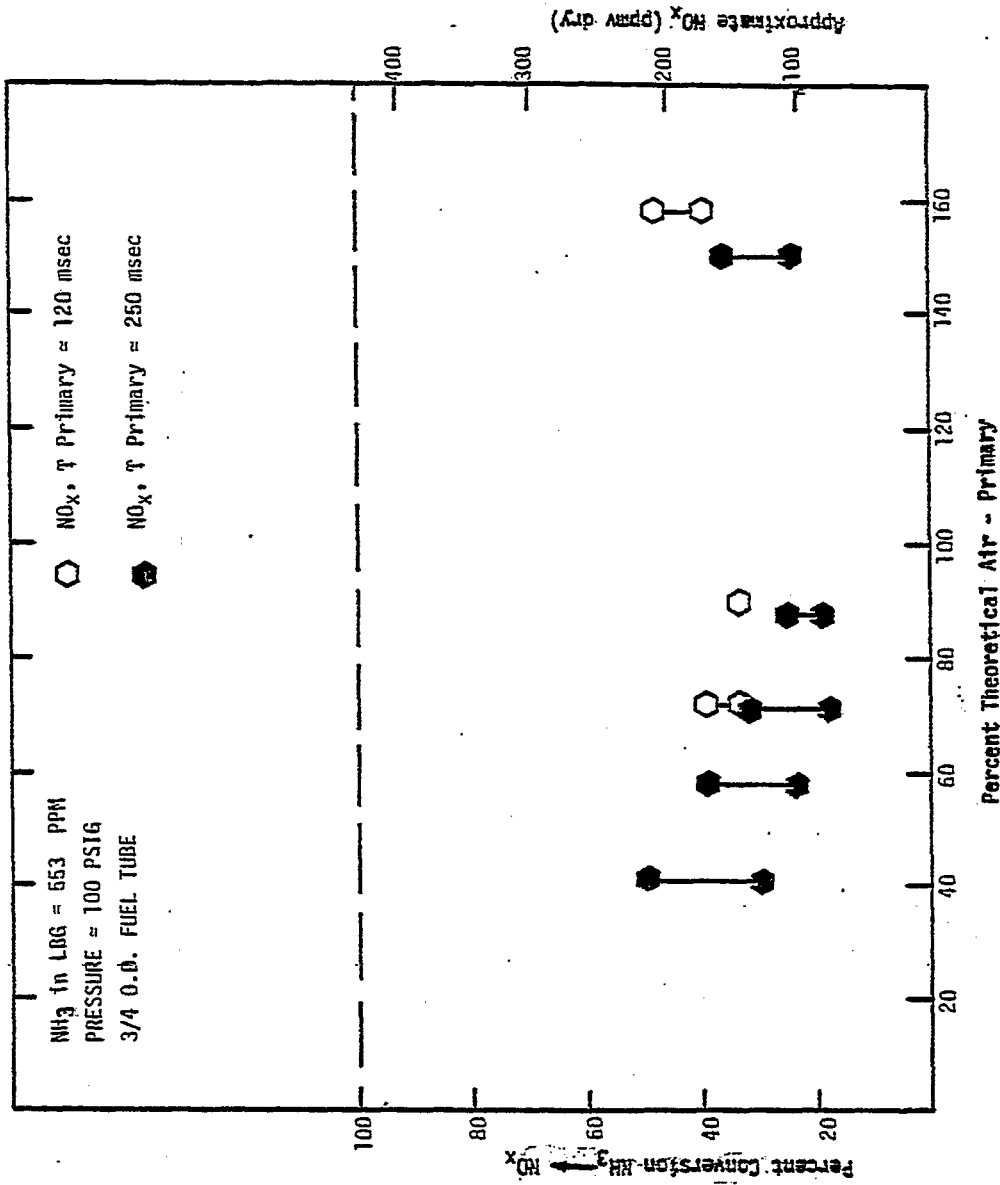Figure 9. Bench-Scale Diffusion Flame: Variable Pressure/Reynolds Number.

Figure 10. Bench-Scale Diffusion Flame: Effect of Residence Time.

conversion with primary stoichiometry for two different primary residence times. Using a large pressure-control orifice, a pressure of 8 atmospheres was achieved at fuel-tube Reynolds numbers around 40,000 and primary residence times around 120 msec. A minimum XN conversion of 34 percent was observed at a primary stoichiometry around 90 percent theoretical air. Using a smaller pressure control orifice, a pressure of 8 atmospheres was achieved at fuel-tube Reynolds numbers around 20,000 and primary residence times around 250 msec. A minimum XN conversion of about 22 percent was observed at a primary stoichiometry around 90 percent theoretical air. For a 553-ppm-doped LBG burned out to 150 percent theoretical air, this XN minimum corresponded to a NO_x concentration of 100 ppm.

Figure 11 shows NO_x concentration as a function of NH_3 in the LBG for rich/lean staged combustion in a diffusion flame and in a platinum/nickel oxide catalytic combustor. For both the catalytic and the diffusion-flame combustors, NO_x emissions increased with increasing fuel nitrogen content, but the increase in NO_x was much less than proportional to the increase in fuel nitrogen content. The 3/8 OD tube diffusion flame, operated at a primary stoichiometry of 76 percent theoretical air and a pressure of 4.4 atmospheres, converted 40 percent of its fuel nitrogen to NO_x at 553-ppm NH_3 in the LBG and had conversions of only 11 percent at a 3800-ppm doping level. The catalyst, operated at a primary stoichiometry of 80 percent theoretical air and a pressure of 2.4 atmospheres, had XN conversions of 16 percent at the low NH_3 doping level and 6 percent at the high doping level. Similar trends, but higher NO_x concentrations, were observed for both combustors in fuel-lean combustion.
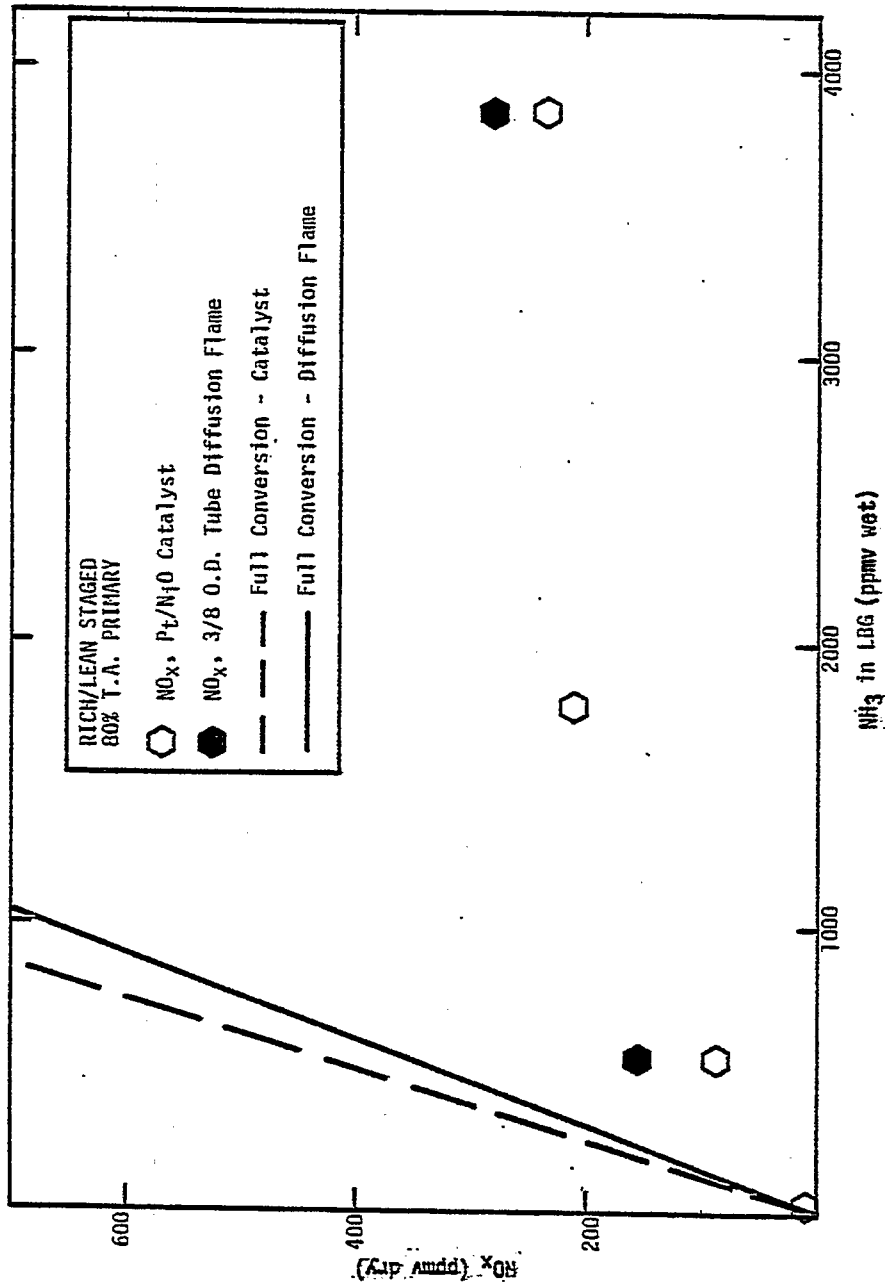
Figure 11. Bench-Scale Reactor Comparison: Effect of Dopant Level.

SECTION 4

LIQUID FUEL - EXCESS AIR RESULTS

PETROLEUM LIQUIDS

To define the influence of fuel composition on total and fuel NO_x emissions, each oil was tested under similar conditions in the tunnel furnace. Fuel NO_x formation was determined by substitution of the combustion air with a mixture of argon, oxygen, and carbon dioxide. The argon replaced the nitrogen, thereby eliminating thermal NO_x formation and the CO_2 provided the proper heat capacity so that flame temperatures were matched. Total and fuel NO_x emissions were measured with an air preheat level of 405 $\pm5^\circ K$ and an atomization pressure of 15 psig. Figure 12 presents a composite plot for total and fuel NO_x (defined by argon substitution) as a function of weight percent nitrogen in the fuel for a wide range of petroleum and blended distillate fuels. In Figure 12 the various symbols represent different base fuels (see Table 1 for symbol key). Those symbols shown with a line refer to distillate or residual fuels doped with pyridine or thiophene. It can be seen that both total and fuel NO_x increase with increasing fuel nitrogen content, and that total fuel nitrogen level is the dominant factor controlling fuel NO_x formation in this system. The form of the nitrogen does not appear to significantly influence fuel NO formation under excess air conditions, as doping with a volatile nitrogen compound (pyridine) resulted in NO_x emission similar to that from a less volatile residual oil of the same nitrogen content. Since the data is for a system where very fine oil droplets (approximately 25 micron mean diameter) are well-dispersed in the oxidizer under hot fuel-lean conditions, it is not surprising that fuel NO_x emissions are somewhat higher than those achieved in practical systems.
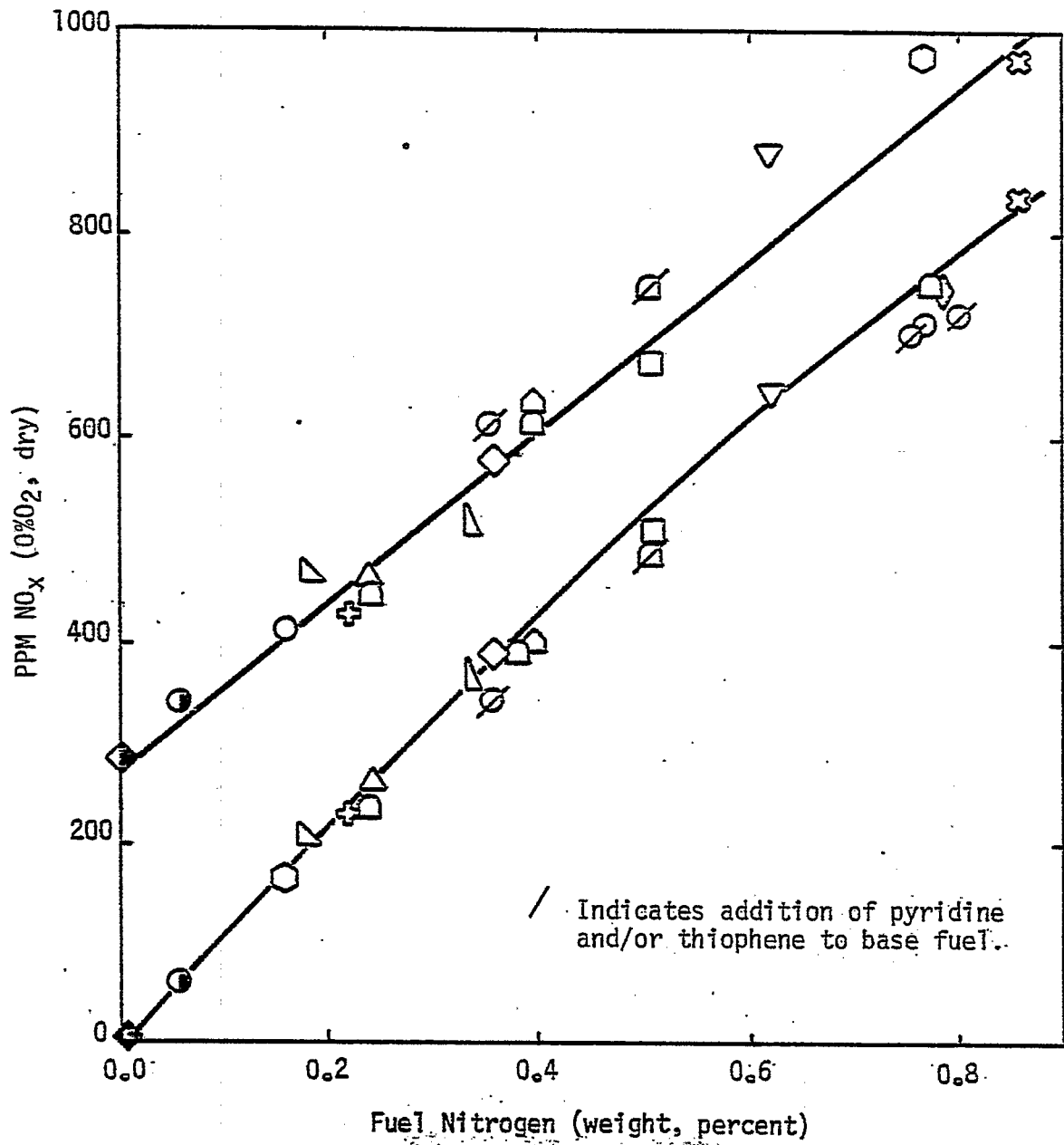
Figure 12. Total and Fuel NO_x Emissions From Pure and Doped (pyridine and thiophene) Petroleum Fuels Tunnel Furnace.

184

ALTERNATIVE FUELS

Figure 13 presents a composite plot of total and fuel NO_x for the range of petroleum fuels together with alternative fuels and mixtures. The Paraho shale was mixed with the same low sulfur oil used by Mansour (19). Synthoil could not be pumped without blending and the results presented in Figure 4 refer to 80 and 90 percent Synthoil blends with distillate oil. The SRC-II blend refers to a mixture of SRC-II and the donor solvent. Under the conditions tested, fuel NO_x emissions increase approximately linearly with increasing fuel nitrogen and it can be seen that the fate of fuel nitrogen in alternative fuels is similar to that in petroleum-derived fuels. Figure 14 presents the fuel NO_x data plotted as a percentage of the fuel nitrogen converted to fuel NO_x. For low fuel nitrogen contents, the conversion decreases rapidly (from greater than 90 percent) as fuel N increases. Eventually, however, the conversion becomes almost independent of fuel nitrogen content; hence, the linear dependence shown in Figure 13.

The absolute level of the fuel N conversion can be influenced by altering the fuel/air contacting and/or the fuel atomization (2), but the results obtained in this study suggest that fuel nitrogen is the only first-order fuel composition parameter controlling NO_x formation in fuel-lean flames. This conclusion applies to petroleum-, coal-, and shale-derived liquid fuels. However, there appear to be second-order effects where the volatility of the fuel nitrogen compound does have an influence upon fuel NO_x formation. Comparison of the data for the fuels with fuel nitrogen content of approximately 0.24 percent indicates that the highest conversion is achieved with a shale-derived distillate fuel with a large volatile nitrogen fraction.
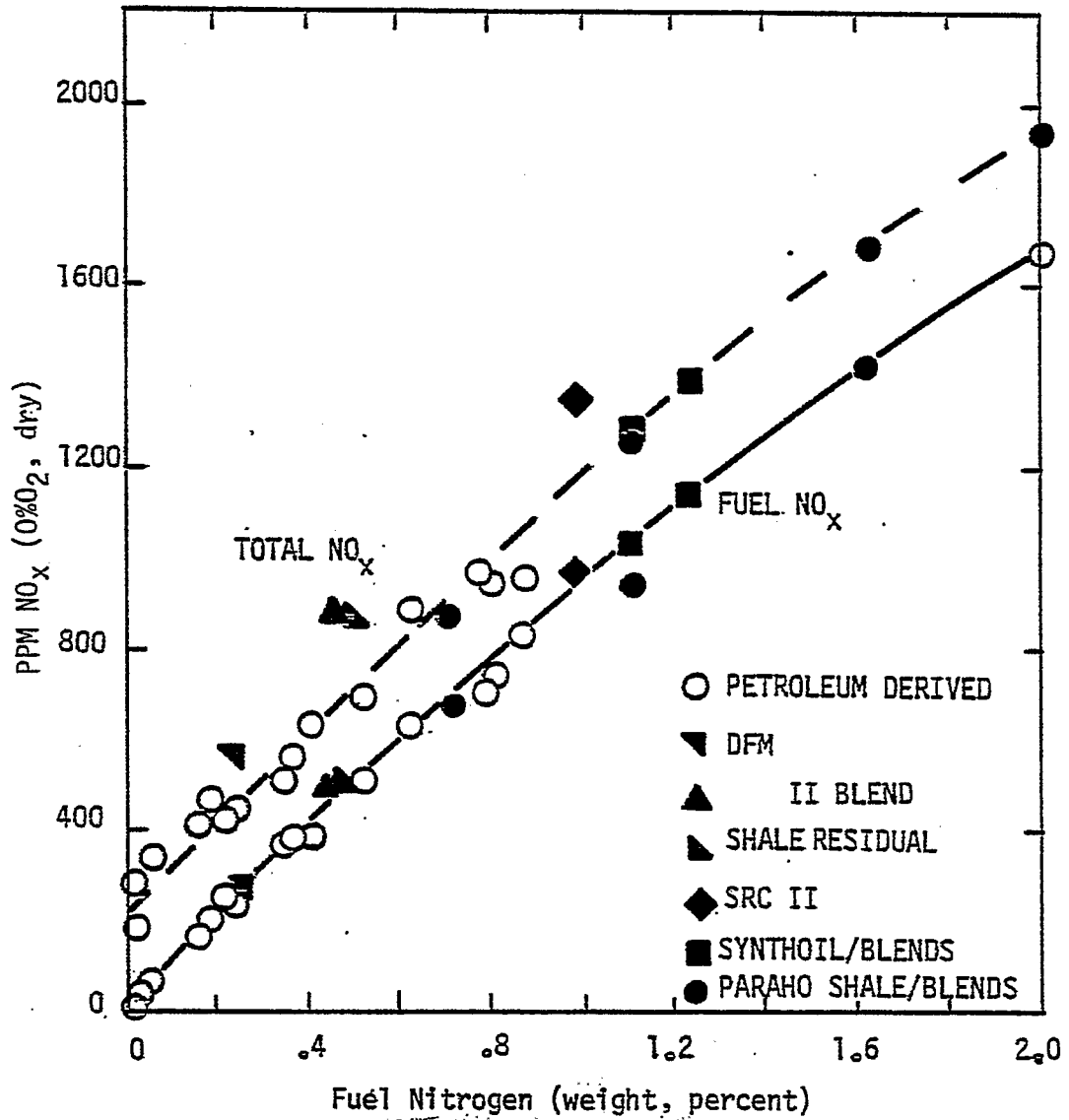
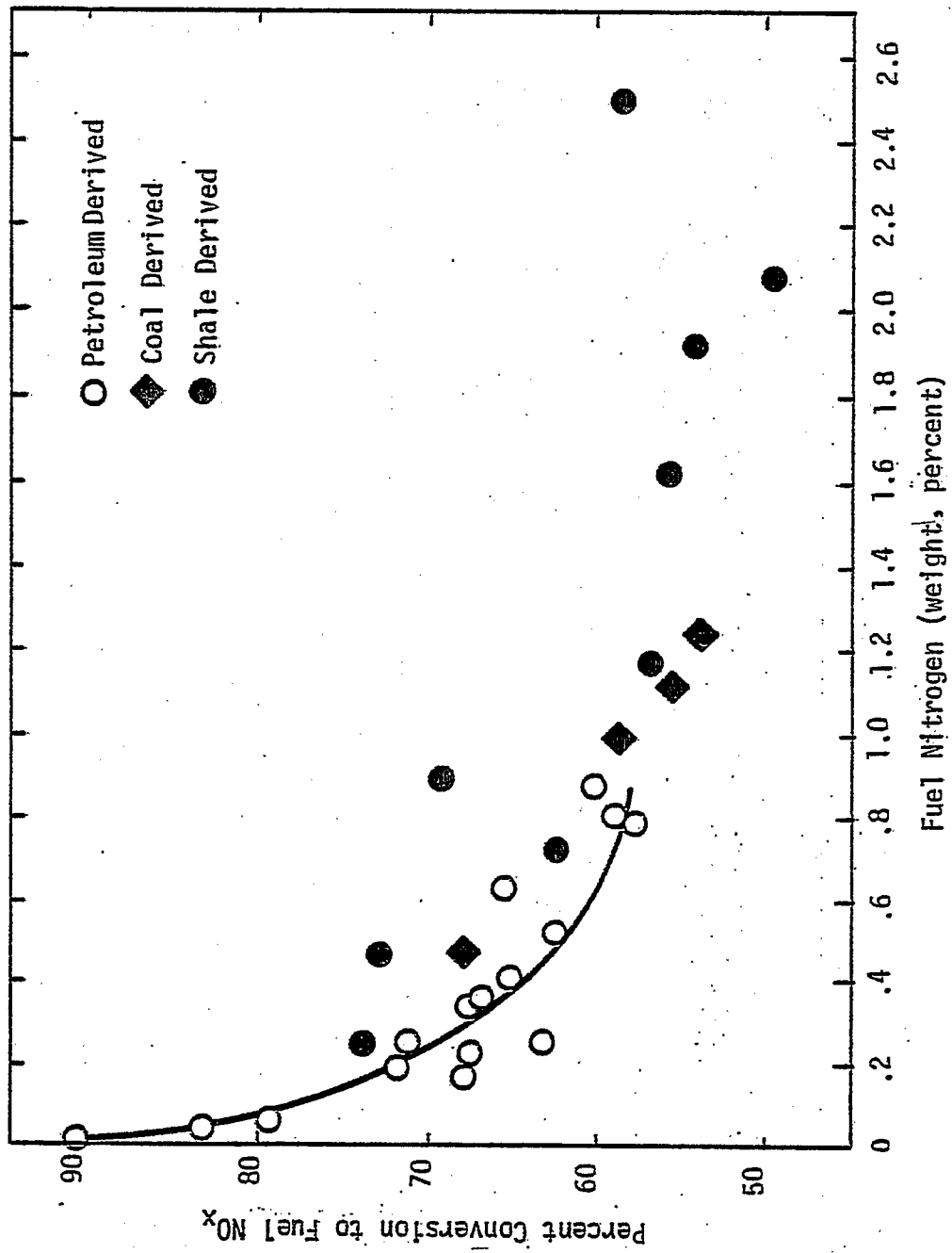Figure 13. The Effect of Fuel Nitrogen Content on Total and Fuel NO_x (5 percent excess oxygen).

Figure 14. Fuel Nitrogen Conversion – Comparison of Alternate and Petroleum-Derived Fuels.

187

SECTION 5

LIQUID FUELS STAGED COMBUSTION RESULTS

POTENTIAL FOR NO_x CONTROL

Staged combustion, i.e., the operation of a combustion system in which the fuel originally burns under oxygen-deficient conditions, provides the most cost-effective control techniques established to date for reducing fuel NO_x. Figure 15 shows the influence of primary zone stoichiometric ratio on total NO_x emissions for two coal-derived and two shale-derived liquids under staged combustion conditions and 3 percent overall excess O_2. All the data in Figure 15 were obtained in the tunnel furnace with ultrasonic atomization and with a first-stage residence time of approximately 800 ms. As the primary zone becomes more fuel-rich, NO_x emissions decrease dramatically to a minimum and then increase again. This trend is in agreement with previously-reported data on petroleum fuels (21).

FUEL CHEMISTRY

First Stage Stoichiometry

In an effort to better understand the mechanisms of NO formation under staged combustion conditions, the original furnace was modified to allow in-flame sampling of the XN (NO, HCN, NH_3) species and cooling of the first-stage and/or second-stage combustion products, as illustrated in Figure 16. A "radiation shield" (choke) was installed near the top of the furnace to minimize the effects of downstream changes on the fuel vaporization zone. A secondary air injection ring and cast refractory choke were installed at 41 in. to insure isolation of the first stage. Variable cooling was achieved by insertion of multiple stainless steel water-cooling coils.

Figures 17, 18, and 19 show typical results of the detailed in-flame measurements made at the exit of the first stage for a distillate oil (D1-Alaskan
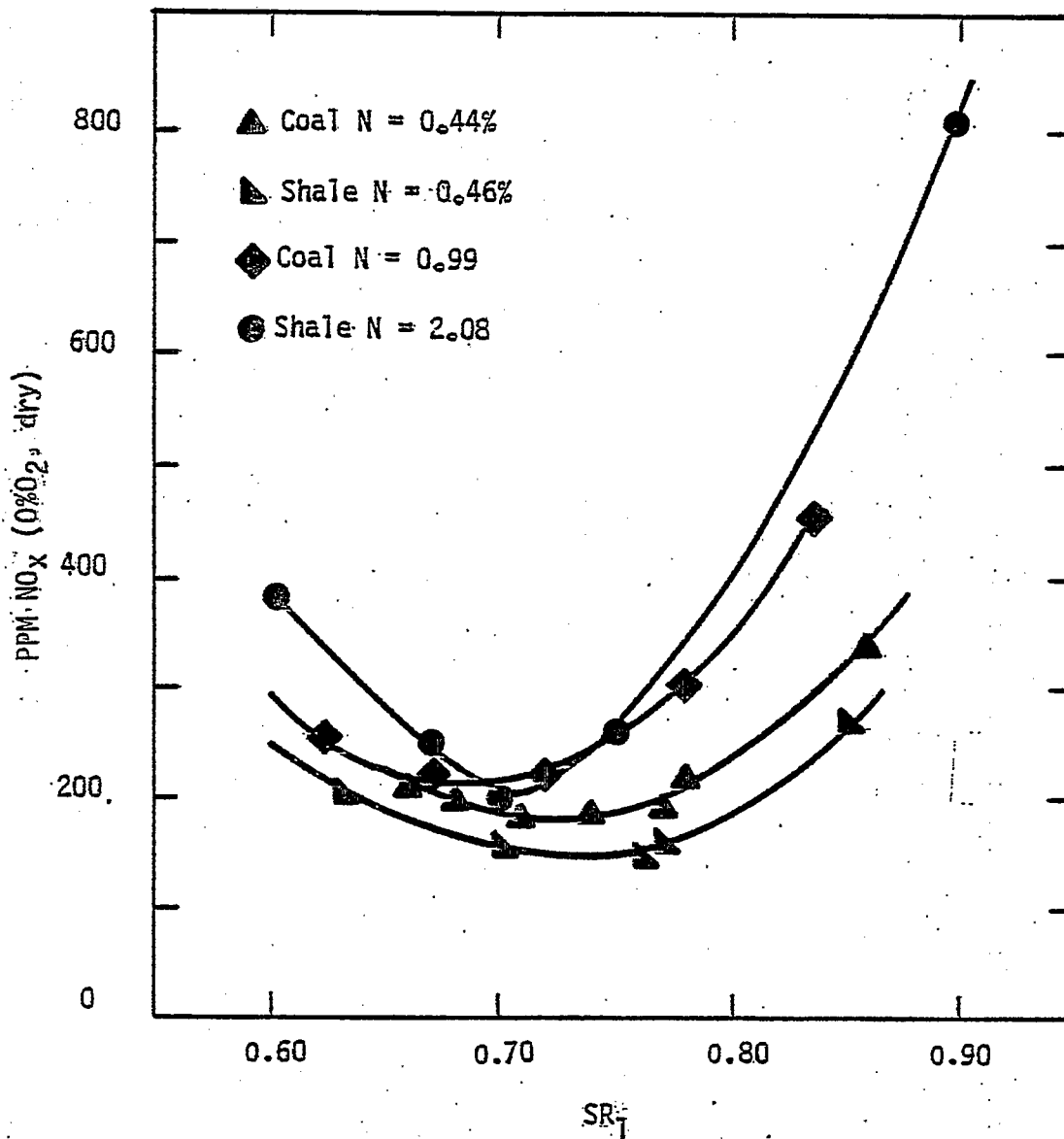
Figure 15. Minimum NO_x Levels Achieved With Alternate Fuels
(tunnel furnace primary zone residence time .83 sec)..

189

Cast Refractory Choke Section

Secondary Air Injector

Cooling Coils

Atomizing Air

Oil

Combustion Air

0
6
12
18
24
30
36
42
48
54
60
66
72
78
84
90

Nozzle Position

Figure 16. Modified Tunnel Furnace.

190

Figure 17. Detailed In-Flame Species Measurements With Alaskan Diesel Oil (D1).

191

Figure 18: Detailed In-Flame Measurements
With Kern County Crude Oil (R14).

Figure 19. Detailed In-Flame Measurements With +850°F Shale Fraction (A8).

diesel), a high nitrogen residual oil (R-14-Kern County, California) and an alternative liquid fuel (A8-+850°F shale fractio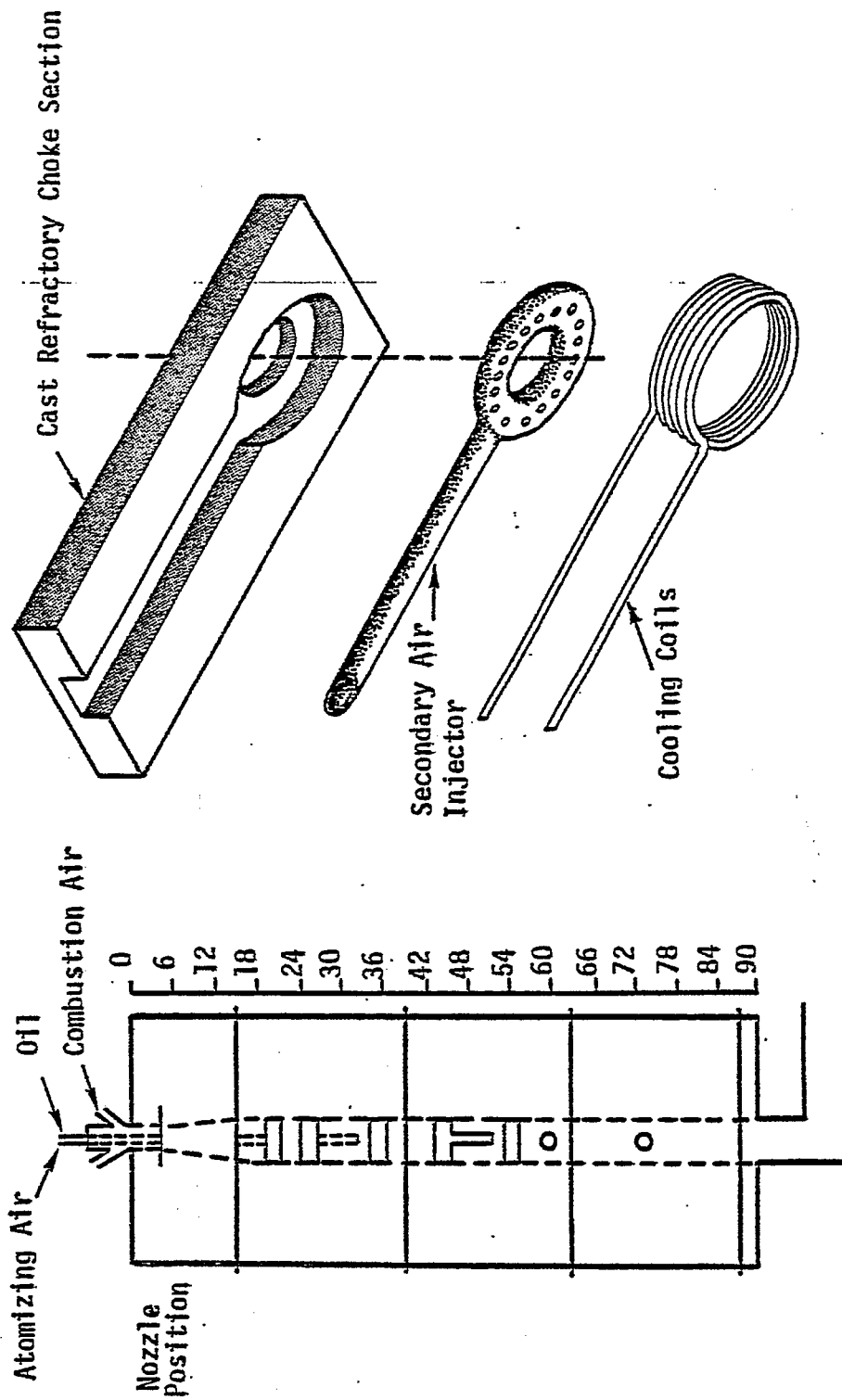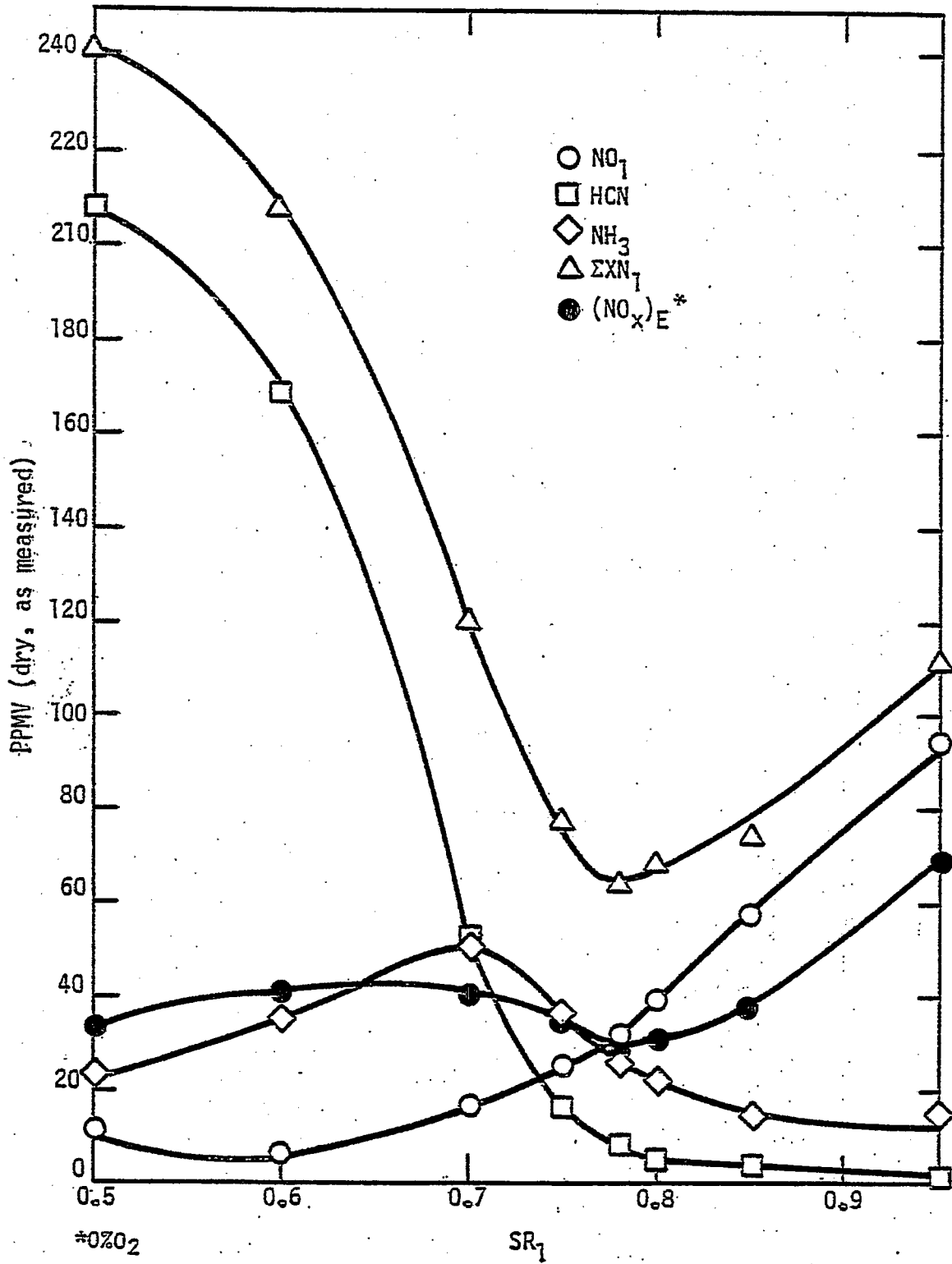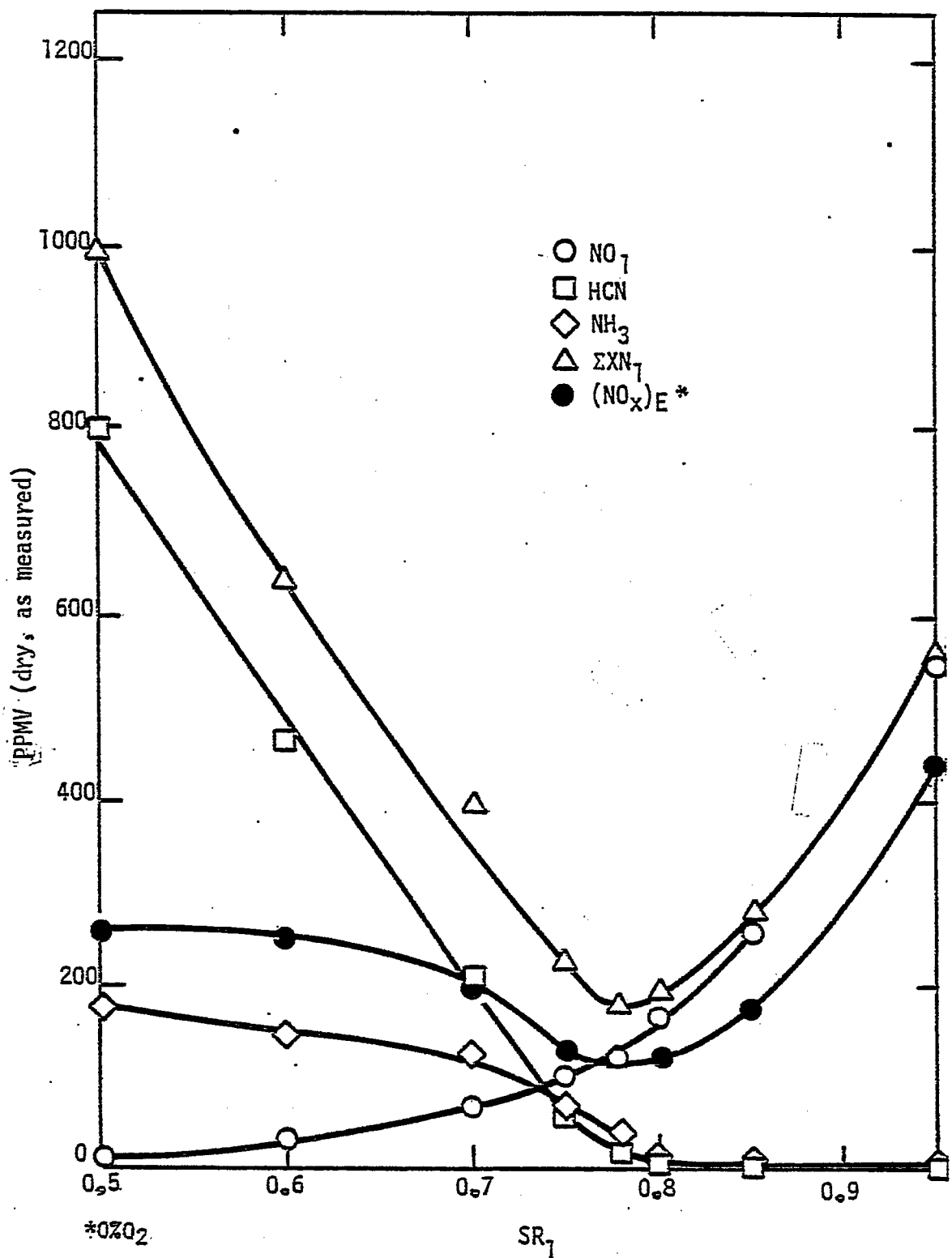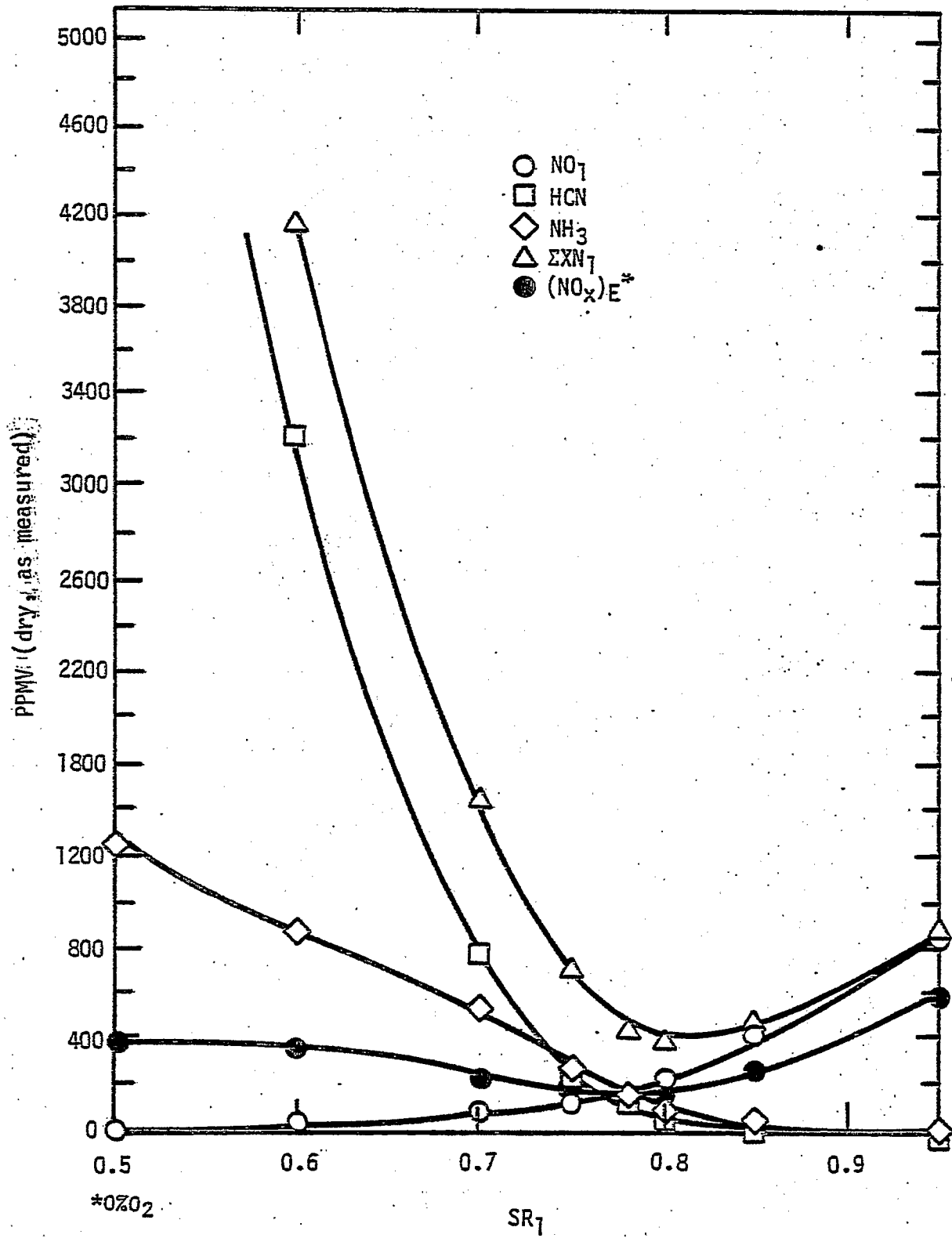n). These measurements were made on the centerline of the furnace at a distance of 104 cm (approximately 630 msec) from the oil nozzle. Detailed radial measurements indicated that the concentration profile was essentially uniform at this location. All of the in-flame data are reported on a dry, as-measured basis. After each in-flame measurement, second-stage air was added at 107 cm and exhaust NO_x measurements were also made (shown on a dry, 0% O_2 basis). In general, decreasing the first-stage stoichiometric ratio reduced the NO concentration leaving the first stage. However, below a stoichiometric ratio of approximately 0.8 significant amounts of NH_3 and HCN were measured. Thus, there exists a minimum in exhaust NO_x concentrations because of a competition between decreased first-stage NO and increased oxidizable nitrogen species such as HCN. Figures 18 and 19 indicate that the petroleum-derived oil (0.83 percent N) and the heavy shale liquid (2.49 percent N) produce large amounts of HCN. In addition, both fuels exhibited a minimum in TFN at a first-stage stoichiometry of approximately 0.8.

Data for the Alaskan diesel oil (Figure 17) also show the presence of much smaller but significant concentrations of HCN and NH_3, although this fuel is essentially nitrogen-free. Total conversion of the fuel nitrogen would produce 21 ppm TFN at SR_1=0.7. This confirms previous work (10-12) which demonstrated that reactions involving hydrocarbon fragments and N_2 or NO can produce HCN.

Hydrocarbons

The rapid increase in HCN concentration below SR_1=0.8 was accompanied by an increase in hydrocarbon content of the partially oxidized combustion products. Figure 20 summarizes the in-flame hydrocarbon measurements for the Alaskan Diesel (D1), three petroleum-derived residual oils (Indonesian-R4, Alaskan-R9, Kern County-R14), three alternative liquids (SRC-II heavy distillate-A9, crude shale-A3, heavy shale fraction-A8) and methane containing 0.75 weight percent nitrogen as NH_3 (∅). Hydrocarbon concentrations correlate well on the basis of first-stage stoichiometry. At very low stoichiometric ratios the distillate oil (◆) and CH_4/NH_3 produced slightly higher hydrocarbon concentrations than the heavier liquid fuels.
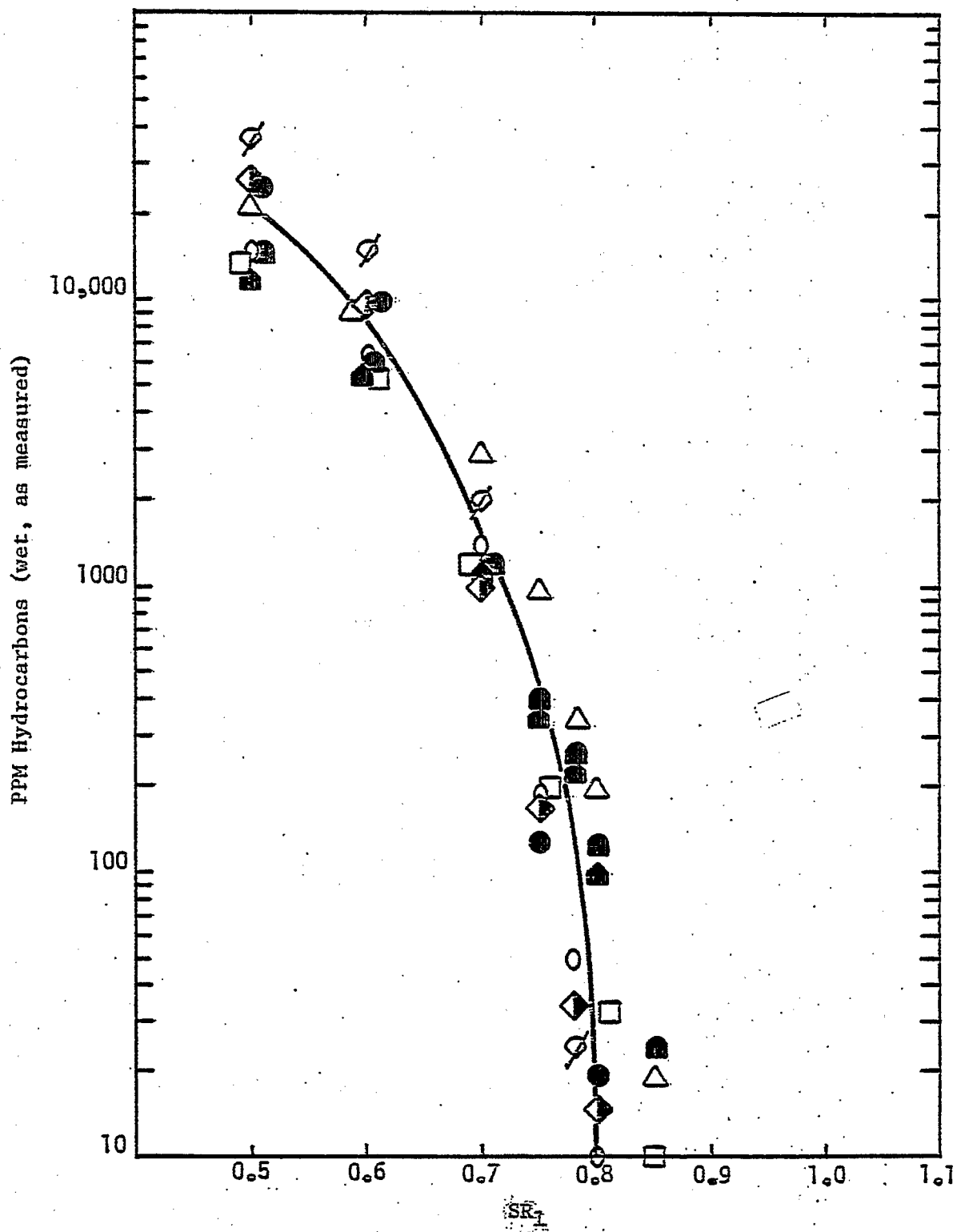
Figure 20.   First-Stage Hydrocarbon Production.

XN Distribution

Figure 21 shows typical results on the percentage of the original fuel nitrogen existing as either NO, NH_3 or HCN at various stoichiometric ratios for four fuels. Above $SR_1 = 0.8$, NO was the dominant TFN species; at lower stoichiometries HCN dominated with all fuels tested except the CH_4/NH_3. Axial profiles with the liquid fuels indicate that near $SR_1 = 0.8$, significant amounts of NH_3 may be formed early in the rich zone but they decay rapidly. These data are in strong contrast to similar results obtained with pulverized coal (20) which indicate that the preferred TFN species is a strong function of coal composition.

In general, both the alternate and petroleum-derived liquid fuels behaved very similarly with the exception of the Kern County, California crude (R14). It produced less HCN under rich conditions, and this tendency cannot be readily associated with common fuel properties. Hydrocarbon and nitrogen distillation data indicated that in terms of equilibrium volatile evolution the Kern County fuel is intermediate among the liquids tested. The Indonesian oil was the lightest of the liquid fuels and it produced the highest TFN concentration at the minimum $(SR_1 = 0.8)$.

SECOND-STAGE NO_x FORMATION

Exhaust NO_x emissions in a staged combustor result from conversion of TFN exiting the first stage and any thermal NO_x production during burnout. Thermal NO_x production was not considered to be significant in this study because changes in heat extraction in the burnout region had almost no effect on final emissions. Figure 22 shows exhaust NO_x emissions as a function of total fixed nitrogen in the first stage at stoichiometries between 0.5 and 0.8 for all fuels. The form of this correlation can be compared with that presented in Figure 12 for excess air conditions since the second stage burnout can be considered an excess air flame. Exhaust emissions increase with increasing oxidizable nitrogen content, but the conversion efficiency decreases as the TFN concentration increases. There are three possible explanations for the data scatter shown in Figure 22: (1) TFN is not indicative of the oxidizable nitrogen compounds that are leaving the first stage; (2) TFN conversion in the burnout zone is dependent upon the form of the TFN;
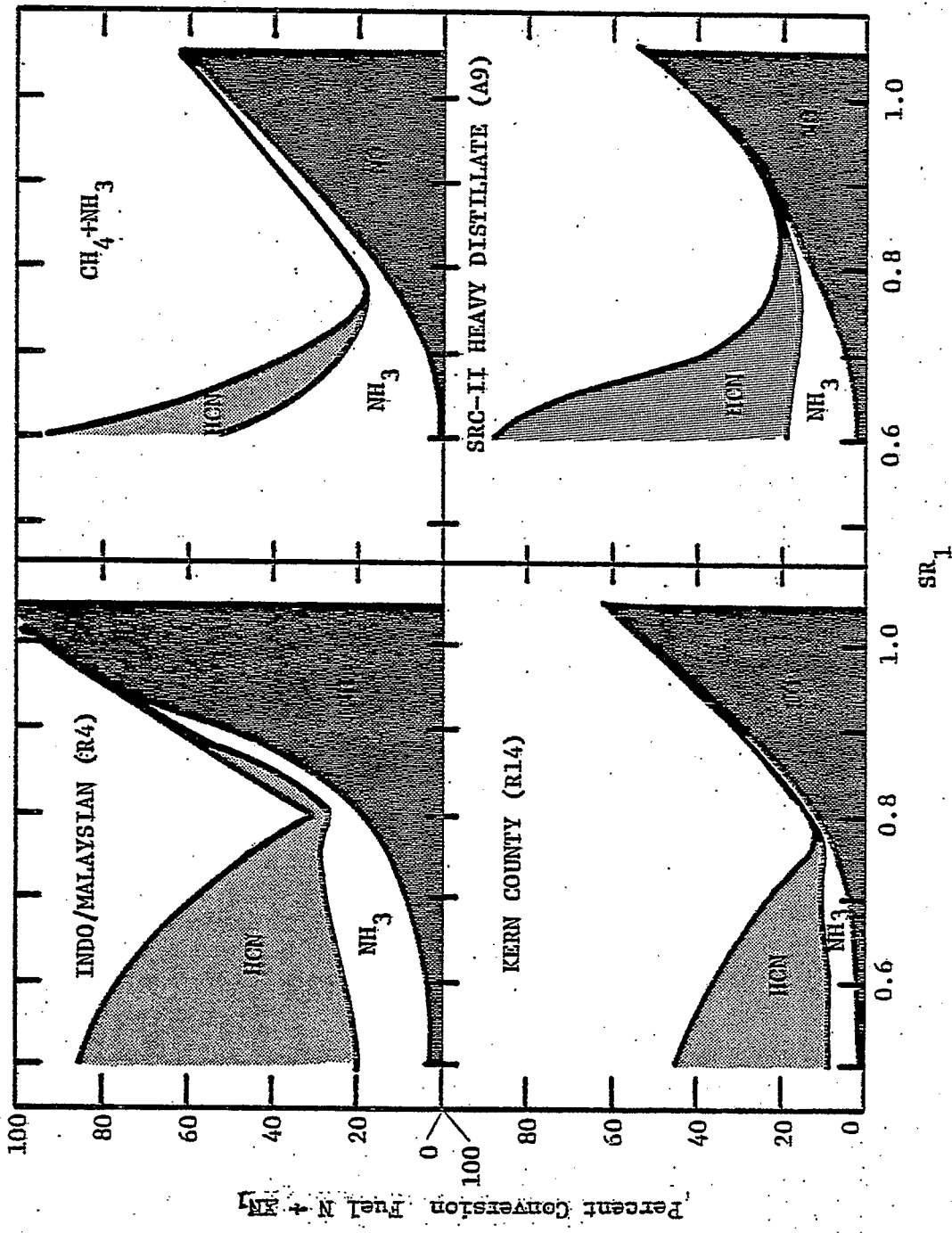
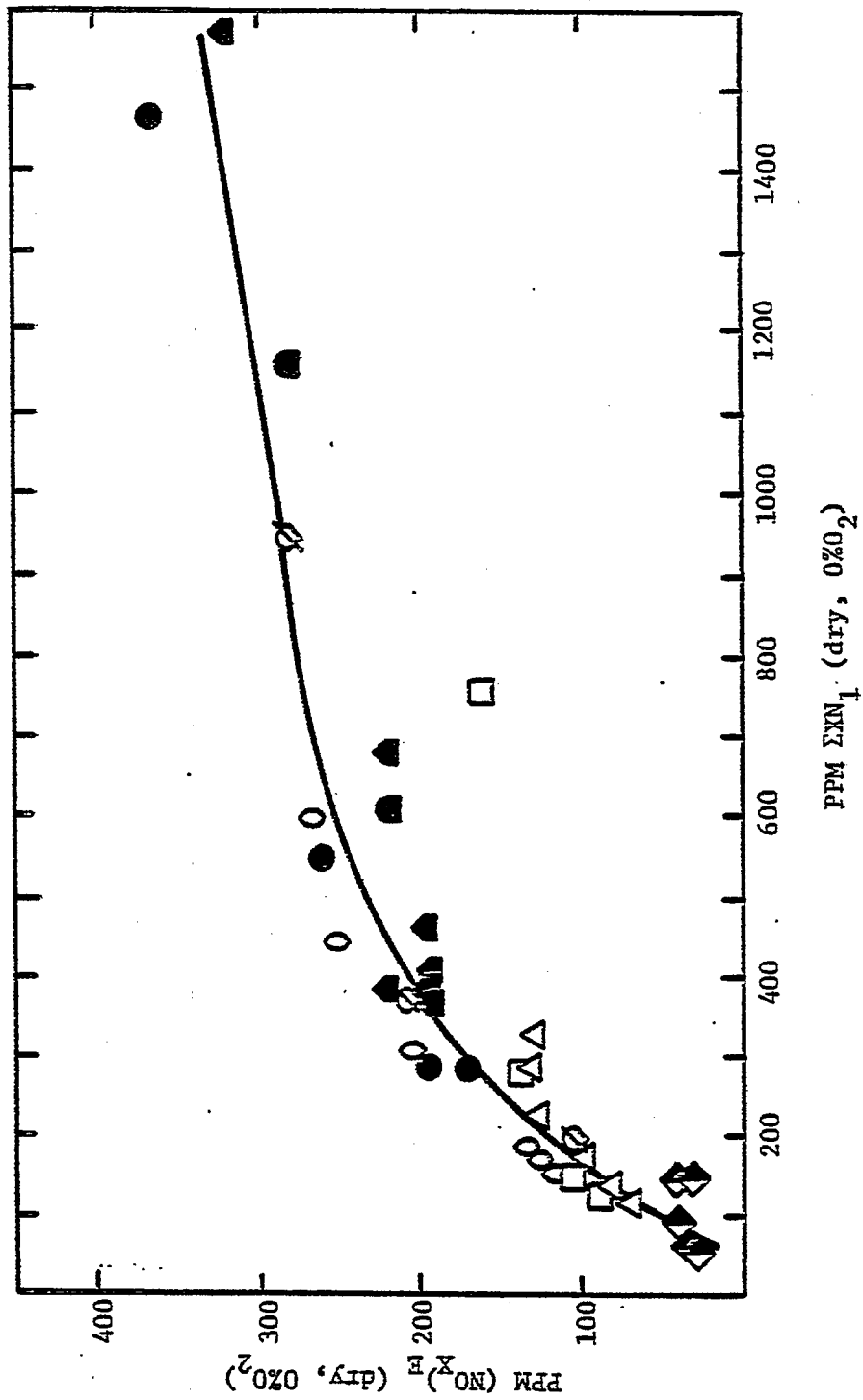Figure 21. Distribution of First-Stage XN Species for Alternative and Petroleum-Derived Fuels.

197

Figure 22. Exhaust NO_x Versus TFN at the Exit of the First Stage.

and (3) TFN conversion is also dependent upon the oxidation of the partial products of combustion at the exit of the fuel-rich zone.

IMPACT OF THERMAL ENVIRONMENT

The TFN concentrations shown in Figure 21 are in excess of equilibrium levels and Sarofim and co-workers (25) have suggested that increasing the temperature of the primary zone would prove beneficial. The results presented in Figure 23 were obtained with the shale crude (A3) to demonstrate the impact of first- and second-stage heat removal on the fate of fuel nitrogen. Figure 23a indicates that adding the radiation shield with cooling coils in both the first- and second-stage (hence, increasing the temperature of the vaporization zone) reduced the minimum NO_x emissions and shifted the optimum stoichiometry more fuel-rich. Figure 23b shows that removing the water cooling coils from the first stage reduced the exhaust emissions. Removing the second stage coils did not alter the minimum level; however, it did shift the minimum SR more fuel-rich. Thus, the optimum thermal environment has a high temperature vaporization zone, a hot, rich hold-up zone, and a cooled second stage (Figure 23c).

The axial profiles (22) provide an explanation for this shift in the minimum emission levels. Heat extraction in the first stage impacts the rate of decay of TFN. Under cold conditions, both NO and HCN essentially freeze, whereas without heat extraction the initial rate of decay for all three species is much faster leading to low TFN concentrations at the exit of the fuel-rich first stage. It should be noted that heat extraction also affects the rate of CO oxidation.
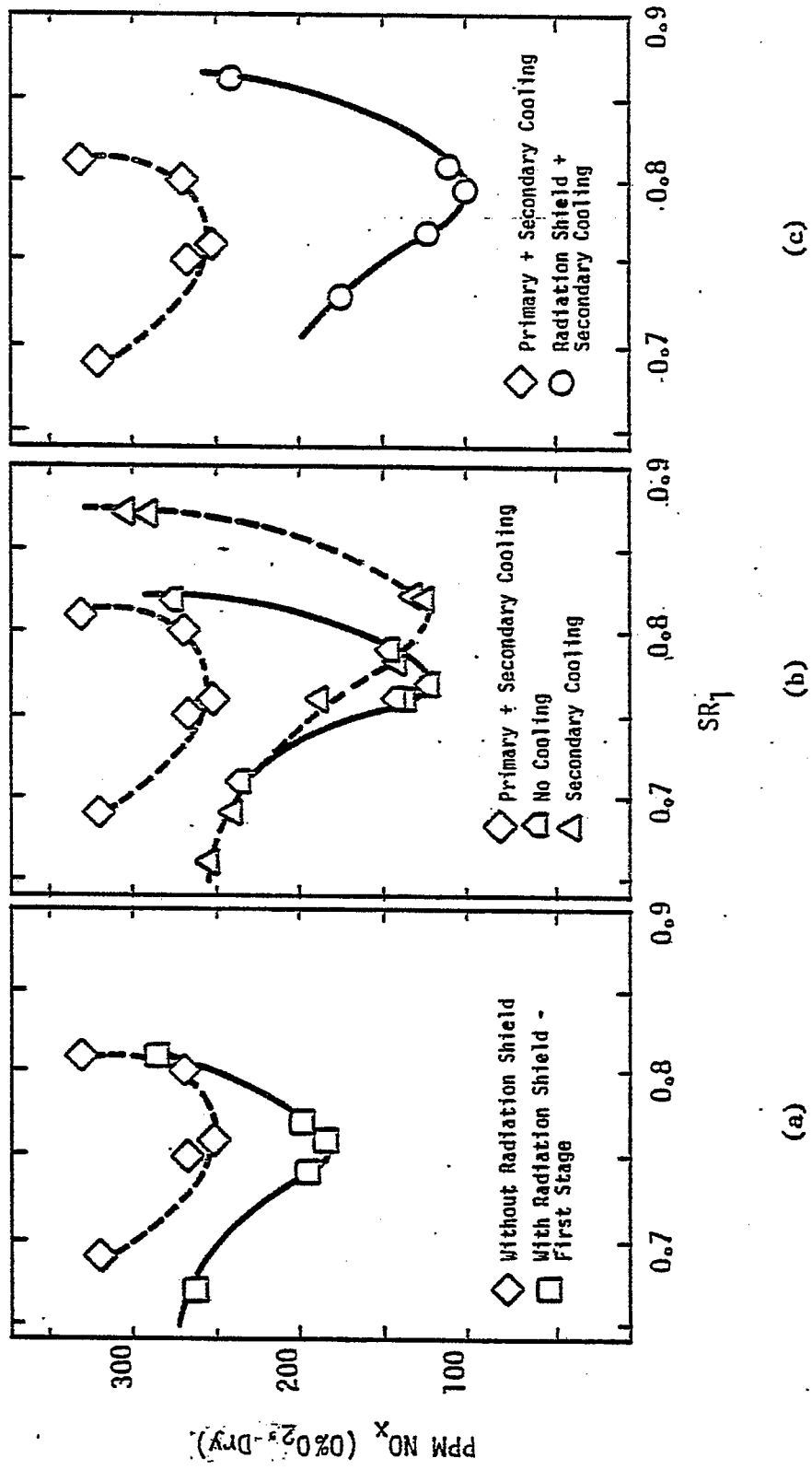
Figure 23. Influence of Heat Extraction Profile in the First and Second Stage Upon Exhaust NO_x Emissions.

200

SECTION 6

CONCLUSIONS

A rich/lean series staged combustor with a platinum/nickel oxide primary was the most promising low-NO_x combustor investigated with LBG. It had low conversions of fuel nitrogen to NO_x over a wide range of fuel-rich primary stoichiometries. Thus, it could be operated rich enough to maintain the adiabatic flame temperature relatively cool, prolonging the life of the catalyst. However, catalyst coated ceramics are often short-lived due to loss of activity of the coating and structural problems of the support caused by thermal shock. During the course of the bench-scale experiments there was a great change in the appearance of the Pt/NiO catalyst. A green coating formed on the surface. Also, the zirconia honeycombs became quite fragile after repeated thermal cycling, especially the fine-cell downstream monolith which was almost completely destroyed in the final experiments. Further investigation is necessary of catalyst aging and of pressure and throughput effects under optimized combustor conditions before a catalytic combustor could be considered a serious candidate for a gas turbine combustor.

A rich/lean series staged combustor with a diffusion flame primary also had low conversions of fuel nitrogen to NO_x. Primary stoichiometry and residence time had the most significant effects on fuel nitrogen conversion. Minimum NO_x emissions were achieved at primary stoichiometries around 90 percent theoretical air for long primary residence times (250 msec or longer). Pressure and Reynolds number had little effect on NO_x in a staged diffusion flame, while an increase in the hydrocarbon content of the LBG caused a slight increase in NO_x emissions. Combustion of a hydrocarbon-free LBG was not tested on the bench scale, but laboratory-scale tests indicated that the absence of hydrocarbons in the fuel could cause a significant reduction in NO_x emissions.

A lean unstaged diffusion flame produced higher NO_x emissions than the rich/lean staged diffusion flame. However, because of its simplicity, it remains an attractive low NO_x combustor concept. The influence of Reynolds number on NO_x levels in the lean flame suggests that NO_x emissions could be lowered by utilizing larger fuel tubes, perhaps approaching the levels achieved by the staged diffusion flame.

It is planned to investigate other combustor configurations including a premixed backmixed simulated stirred reactor and a combination diffusion flame/ catalyst hybrid combustor. The zero dimensional stirred reactor is easy to model. It will provide experimental feedback for the fuel nitrogen processing kinetics code to be used in future prototype combustor design. The hybrid system will input low ΣXN containing fuel-rich diffusion-flame exhaust into a Pt/NiO cleanup catalyst prior to secondary burnout.

The results of the bench-scale studies on the influence of liquid fuel properties and thermal environment on NO_x formation indicate that:

- With liquid fuels, fuel nitrogen content is the primary composition variable affecting fuel NO formation. NO_x emissions increase with increasing fuel nitrogen. Alternative liquid fuels correlate with the high-nitrogen petroleum oils.

- Staged combustion dramatically reduces both fuel and thermal NO_x formation. Minimum emissions occur at a primary zone stoichiometric ratio between 0.75 and 0.85 depending on the combustion conditions.

- First-stage stoichiometry determines the dominant TFN species. Below $SR_1=0.8$ HCN is the dominant species, and above $SR_1=0.8$, NO is the dominant species. NH_3 concentrations at the first-stage exit generally accounted for less than 20 percent of the fuel nitrogen.

- Exhaust NO_x emissions are directly related to the TFN concentration at the first-stage exit. NO_x control for high-nitrogen fuels is most effective when a rich primary zone is held at an optimum stoichiometry to minimize the TFN concentration. This concentration is further minimized by increasing the temperature of the fuel-rich zone.

Figure 24 summarizes the impact of fuel-bound nitrogen content on minimum emissions observed under staged combustion conditions and the associated TFN. Under optimum staged conditions NO_x emissions (and TFN) correlate well with total fuel nitrogen content. Only the SRC-II heavy distillate (▲) exhibited unusually high emissions and this was the direct result of a high TFN yield. These results suggest that NO_x emissions resulting from the combustion of coal- or shale-derived liquid fuels can be controlled in a cost-effective manner by modification to the combustion process. Low-NO_x combustors can be designed which are tolerant to wide ranges in fuel-bound nitrogen content. Thus, the production of alternate fuels should be optimized without regard for the reduction of fuel nitrogen content as a method of controlling NO_x emissions from stationary sources.
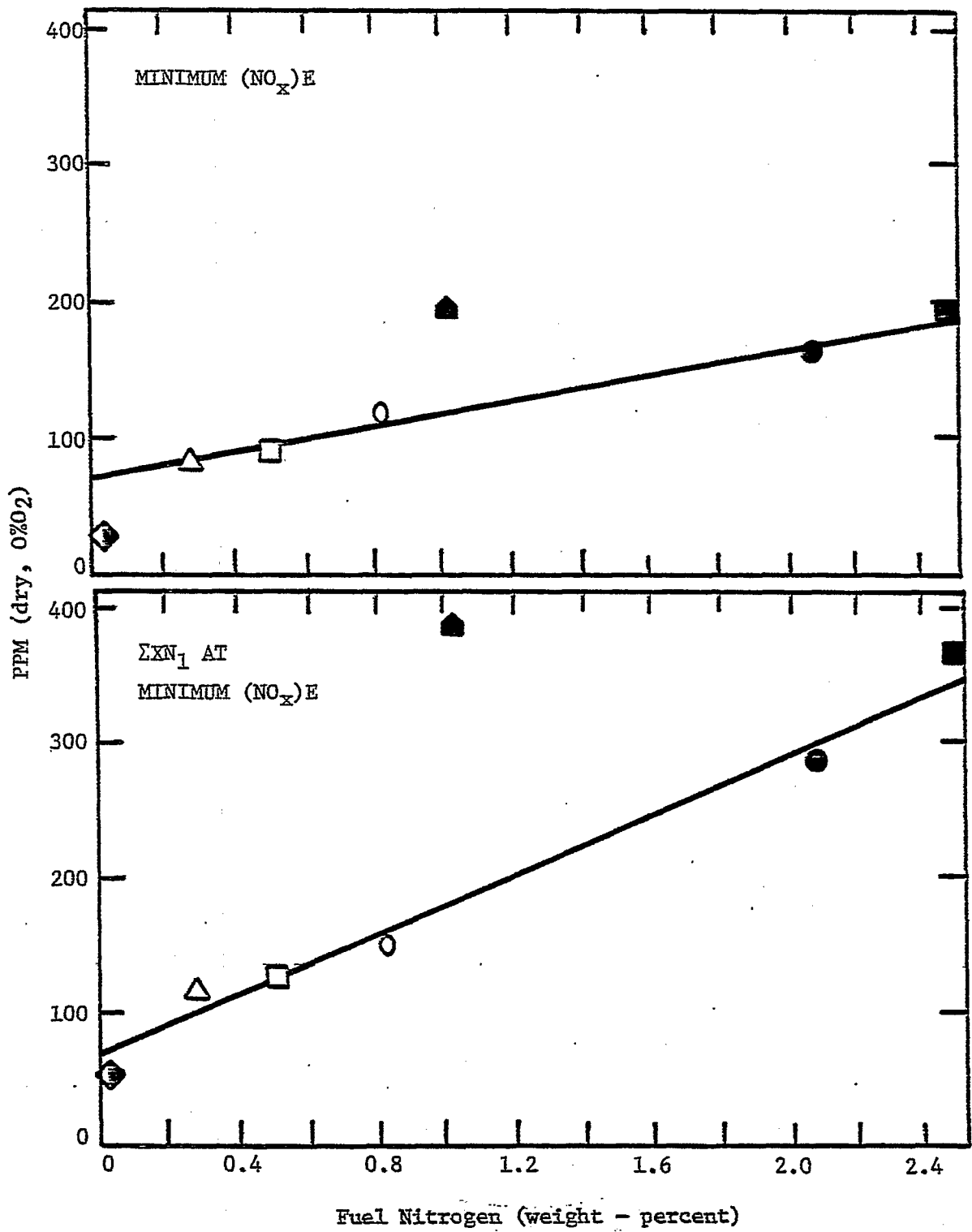
Figure 24. The Effect of Fuel-Bound Nitrogen Content on
Exhaust NO_x and TFN in the Primary Zone (ΣXN_1)
($SR_1 = 0.78$, 3% overall excess O_2).

REFERENCES

1. Mansour, M. N. and M. Gerstein. Correlation of Fuel Nitrogen Conversion to NO_x During Combustion of Shale Oil Blends in a Utility Boiler. In: Proceedings of Symposium on Combustion of Coal and Synthetic Fuels, American Chemical Society, March 1978.

2. Heap, M. P. et al. Control of Pollutant Emissions from Oil-Fired Package Boilers. In: Proceedings of the Stationary Source Combustion Symposium, EPA-600/2-76-1526, NTIS, Springfield, Virginia, 1976.

3. Bowman, C. T. Kinetics of Nitric Oxide Formation in Combustion Processes. In: Proceedings of Fourteenth Symposium (International) on Combustion, The Combustion Institute, Pittsburgh, Pennsylvania 1973.

4. Martin, G. B. and E. E. Berkau. An Investigation of the Conversion of Various Fuel Nitrogen Compounds to NO in Oil Combustion. In: Proceedings of AIChE Symposium Series No. 126, 68, 1972.

5. Turner, D. W., R. L. Andrews, and C. W. Siegmund. Influence of Combustion Modification and Fuel Nitrogen Content on Nitrogen Oxide Emissions from Fuel Oil Combustion. In: Proceedings of AIChE Symposium Series No. 126, 68, 1972.

6. Pershing, D. W., J. E. Cichanowicz, G. C. England, M. P. Heap, and G. B. Martin. The Influence of Fuel Composition and Flame Temperature on the Formation of Thermal and Fuel NO_x in Residual Oil FLames. In: Proceedings of Seventeenth Symposium (International) on Combustion, The Combustion Institute, Pittsburgh, Pennsylvania, 1979.

7. Heap, M. P. NO_x Emissions from Heavy Oil Combustion. International Flame Research Foundation Report for Contract 68-02-0202, IJmuiden, Holland, 1977.

8. Malte, P. C. The Behavior of NH and CN in Nitrogen-Doped High Intensity Recirculative Combustion. Paper presented at the WSS Combustion Institute, Berkeley, California, October 1979.

9. Takagi, T., T. Tatsumi, and M. Ogasawara. Nitric Oxide Formation from Fuel Nitrogen in Staged Combustion: Roles of HCN and NH_i. Combustion and Flame, 35, 17, 1979.

10. Fenimore, C. P. Studies of Fuel Nitrogen species in Rich Flame Cases. In: Proceedings of Seventeenth Symposium (International) on Combustion, The Combustion Institute, Pittsburgh, Pennsylvania, 1979.

11. Haynes, B. S. Reactions of NH_3 and NO in the Burnt Gases of Fuel-Rich Hydrocarbon-Air Flames. Combustion and Flame, 28, 81, 1977.

12. Gerhold, B. W., C. P. Fenimore, and P. K. Dederick. Two Stage Combustion of Plain and N Doped Oil. In: Proceedings of Seventeenth Symposium (International) on Combustion, The Combustion Institute, Pittsburgh, Pennsylvania, 1979.

13. Haebig, J. E., B. D. Davis, E. R. Dzuna. Preliminary Small-Scale Combustion Tests of Coal Liquids. Environmental Science Technology, 10:3, 243, 1976.

14. Muzio, L. J., and J. K. Arand. Small-Scale Evaluation of the Combustion and Emissions Characteristics of SRC Oil. Paper presented at the ACS Fuel Chemistry Symposium, Anaheim, California, March 1978.

15. Downs, W., and A. J. Kubasco. Characterization and Combustion of SRC-II Fuel Oil. EPRI Report No. FP-1028, Palo Alto, California, 1979.

16. Mansour, M. N. Factors Influencing NO_x Production During the Combustion of SRC-II Fuel Oil. Paper presented at the WSS Combustion Institute, Berkeley, California, October 1979.

17. Hersch, S., B. F. Piper, D. J. Mormile, G. Stegman, E. G. Alfonsin, and W. C. Rovesti. Combustion Demonstration of SRC-II Fuel Oil in a Utility Boiler. Paper presented at the ASME Winter Annual Meeting, New York, New York, December 1979.

18. Dzuna, E. R. Combustion Test of Shale Oils. Paper presented at the CSS Combustion Institute, Columbus, Ohio, April 1976.

19. Mansour, M. N., and M. Gerstein. Correlation of Fuel Nitrogen Conversion to NO_x During Combustion of Shale Oil Blends in a Utility Boiler. Paper presented at the ACS Symposium on Combustion, Anaheim, California, March 1978.

20. Chen, S. L., M. P. Heap, R. K. Nihart, D. W. Pershing, and D. P. Rees. The Influence of Fuel Composition on the Formation and Control of NO_x in Pulverized Coal Flames. Paper presented at the WSS Combustion Institute, Irvine, California, 1980.

21. England, G. C., D. W. Pershing, J. H. Tomlinson, and M. P. Heap. Emission Characteristics of Petroleum and Alternate Liquid Fuels. Paper presented at the AFRC NO_x Symposium, Houston, Texas, October 1979.

22. England, G. C., M. P. Heap, D. W. Pershing, R. K. Nihart, and G. B. Martin. Mechanisms of NO_x Formation and Control: Alternative and Petroleum-Derived Fuels. Paper presented at the Eighteenth Symposium (International) on Combustion, The Combustion Institute, Waterloo, Canada, August 1980.

23. Corley, T. L. Development of a Kinetic Mechanism to Describe the Fate of Fuel Nitrogen in Gaseous Systems. Paper presented at the Fifth E.P.A. Fundamental Combustion Research Workshop, Newport Beach, California, 1980.

24. Folsom, B. A., C. W. Courtney, and M. P. Heap. The Effects of LBG Composition and Combustor Characteristics on Fuel NO_x Formation. Paper presented at the ASME Gas Turbine Conference and Exhibit and Solar Energy Conference, San Diego, California, 1979.

25. Sarofim, A.F., J. H. Pohl and B. R. Taylor. Mechanisms and Kinetics of NO_x Formation: Recent Developments. Paper presented at the 69th Annual Meeting AICHE, Chicago, Illinois, 1976.

PROBLEM-ORIENTED REPORT:

UTILIZATION OF SYNTHETIC FUELS:
AN ENVIRONMENTAL PERSPECTIVE

E.M. Bohn, J.E. Cotter, J.O. Cowles,
J. Dadiani, R.S. Iyer, J.M. Oyster

TRW
Energy Systems Planning Division
8301 Greensboro Drive
McLean, VA. 22102

ABSTRACT

This paper discusses the potential environmental problems arising from the refining, transportation, storage and utilization of fuels produced by a synthetic fuels industry. Scenarios defining possible build-up rates for synfuel products from oil shale and coal conversion are developed to scope the magnitude of potential exposures. The market infrastructure for the use of these products is examined and the potential public health risks during the handling, transportation and utilization of these synfuel products is evaluated. Significant issues regarding environmental impacts and the need for regulatory attention are discussed.

SUMMARY

PLANNING FOR SYNFUELS UTILIZATION MUST BEGIN NOW

This document is a preliminary overview intended to broadly sketch out the essential facts of interest to EPA about the utilization of synfuels and their potential environmental impacts. It is also intended to present an overall environmental perspective. A Final Environmental Market Analysis Report will be developed with the purpose of analyzing specific areas of relevance to EPA in greater depth and noting possible EPA activities for mitigating potential environmental impacts of synfuels.

EPA is currently sponsoring projects focussed on the environmental aspects of coal and shale conversion processes. This document deals more with the fate of synthetic fuel products after they leave the plant gate. Future work will be concerned in more detail with the estimated national flow rates and paths of such products and byproducts, their hazards to human health, and the risks of public exposure to these synthetic fuels.

In carrying out its mission of preserving the quality of our natural environment, EPA has the responsibility to keep fully abreast of synthetic fuel developments because a reasoned approach to dealing with the environmental impacts of a synfuels industry requires accurate knowledge about current synfuels processes and commercial applications.

Current trends in the international energy situation are rapidly increasing the probability that a domestic synthetic fuels industry will emerge in the 1980s. Because government incentives and private ventures in the synfuel arena are burgeoning in response to soaring world oil prices and decreasing reliability of oil imports, forecasters are now projecting earlier start dates, faster growth rates, and larger ultimate sizes for such an industry.

Several synfuel technologies are under consideration for commercial production. A wide range of synfuel products are expected to be produced and they will be utilized in a broad category of end uses (reference Table 1). Synfuels products will most likely be used largely as transportation fuel, including gasoline and diesel fuel from refined shale oil and coal conversion processes and jet fuel from refined shale oil. Utility and

Table 1. Synfuels Market Overview

WHAT TECHNOLOGIES PRODUCE SYNFUELS?		WHAT MAJOR PRODUCTS/ BYPRODUCTS WILL THEY MAKE?	WHERE WILL THE PRODUCTS/ BYPRODUCTS BE USED?	WHAT ARE THE RELATIVE POTENTIAL EXPOSURE LEVELS TO THE PRODUCTS?
OIL SHALE:	NUMEROUS RETORTING PROCESSES, INCLUDING TOSCO, PARAHO, UNION, OCCIDENTAL	Syncrude upgraded and refined to yield: LPG Gasoline Jet Fuel Diesel Fuel Residuals Lubricants[a] Waxes[a]	● Commercial and military transportation, including highway vehicles, aircraft, ships ● Utility and industrial boilers ● Commercial and residential heating ● Industrial lubricants	Low for transport of crude shale to refinery; moderate during refining and end use as boiler fuel; increased exposure level when used in transportation sources and space heating.
DIRECT COAL LIQUEFACTION:	SRC-II	LPG Naphtha Fuel Oil SNG[b] Tar Oils[a]	● Utility and industrial boilers ● Commercial and residential heating ● Chemical feedstocks	Low for LPG, SNG, Naphtha, Butane;. Moderate exposure for fuel oils at industrial sites with exposure increasing when used in space heating.
	EXXON DONOR SOLVENT	Propane Butane Naphtha Fuel Oil Solvent[a]	● Utility and industrial boilers ● Commercial and residential heating ● Paint thinners	
	H-COAL	Naphtha Fuel Oil	● Utility and industrial boilers ● Commercial and residential heating	
INDIRECT COAL LIQUEFACTION:	FISCHER-TROPSCH	Gasoline LPG Diesel Fuel Heavy Fuel Oil Medium Btu Gas SNG Tar Oils[a] Phenols[a] Chemical Feedstocks[a] Pesticides[a] Fertilizers[a]	● Commercial and military transportation ● Utility and industrial boilers ● Commercial and residential heating ● Chemical feedstocks ● Agricultural uses	Low for LPG, SNG, and Medium Btu Gas. Moderate exposure when fuels used in transportation sources and boilers. Low to moderate exposure is also estimated when products used as chemical feedstocks.
	M-GASOLINE	Gasoline LPG	● Commercial and military transportation	
	METHANOL	Methyl Fuel Methanol	● Commercial and military transportation ● Chemical feedstocks	
HIGH BTU COAL GASIFICATION:	NUMEROUS PROCESSES, INCLUDING LURGI, COED-COGAS, TEXACO, SHELL-KOPPERS	SNG[b]	● Commercial and residential heating	Very low - similar to current distribution of natural gas.
MEDIUM/LOW BTU COAL GASIFICATION:	NUMEROUS PROCESSES	Medium Btu Gas Low Btu Gas	● Captive fuel use for industrial heating and chemical feedstocks	Very low since it is primarily captive use.

[a] Only representative byproducts are indicated.
[b] Substitute Natural Gas

industrial boilers will utilize the fuel oils produced from coal liquids. High-, medium-, and low-Btu gases from coal will find use in commercial, residential, and industrial heating applications. The products from most synfuel processes will be used as chemical feedstocks in a large variety of industries.

The national environmental impacts of a large-scale synfuels industry could be significant. The environmental concerns of end use, including handling and transport, will have to be investigated in detail. Since there is limited information concerning the end-use exposure effects of synthetic fuel products and by-products, the nature of these future impacts is largely speculative. In fact, since synthetic fuel technology is highly evolutionary, even the composition and amounts of future industrial synthetic fuel products and by-products are not well known.

In this report the term synfuel product refers to primary products of the synfuel industry such as gasoline, high-, medium-, and low-Btu gas, whereas the term by-product has been used to identify secondary useful products that are likely to be produced from synfuels such as plastics, solvents, varnishes, and fertilizers.

A SYNFUEL INDUSTRY IS EMERGING

INCENTIVES FOR SYNFUEL DEVELOPMENT ARE HERE

The primary incentive for synfuels development is the imbalance between domestic supply and demand for petroleum liquids and natural gas. The long-term decline in domestic oil production coupled with increased demand has resulted in a level of oil imports of 9 million barrels per day (MMBPD) of oil or about 50 percent of U.S. consumption. The proven domestic reserves of natural gas are also declining and demand is now being met with increasingly higher priced supplies.

A substantial market for liquid synthetic fuel products and chemical feedstocks is expected by 1990. A recent analysis concludes that about 2.9 Quads of energy or about 1.5 MMBPD will have to be supplied from synthetic liquid fuels (reference Table 2). As indicated in this analysis, use of synfuels is expected to be heavily directed toward transportation. Industry concern over potential interruptions in gas supplies has provided the incentive to develop coal gasification processes to supplement current gas supplies and for use as chemical feedstocks.

It is these considerations, along with the uncertainty inherent in the import supplies and the increasing problem of balance of payments, that now provide the impetus for Federal support for synfuels development. Recent Federal action creating the Synthetic Fuels Corporation (SFC) is aimed at alleviating some of the factors that to-date have discouraged development. The goal of the SFC, with authorized funds for loan guarantees, cooperative agreements, and price supports, is to reduce and share the investment risk of establishing a commercial synfuels industry.

Now, as the U.S. synfuels industry is a developing reality, the EPA will need to initiate close coordination with the SFC. As EPA takes the lead in regulatory approvals, other regulatory agencies will be encouraged to participate. A well organized, coordinated approach on the part of all Federal agencies will be viewed as an added incentive by the developing synfuels industry.

Table 2.   Anticipated Liquid Fuel Products Demand in 1990[a]

	Supplied to Consumers Quads	Petroleum Supplies Quads	Synfuels Quads
Gasoline	14.2	12.7	1.5
Jet Fuel	2.1	1.8	0.3
Kerosene	0.3	0.3	-
Heating Oils	6.4	5.7	0.7
Residual	3.2	3.2	-
Asphalt	1.0	1.0	-
Misc. Product	2.4	2.0	0.4
LPG	0.7	0.7	-
	30.3	27.4	2.9

a.Coal Technology Market Analysis, ESCOE, January 1980.   Assumes
   U.S. refineries will operate with the same mix as 1978.

b 1 Quad/yr = 0.5 MMBPD

SYNFUELS UNDER CONSIDERATION

The term "synfuels" has become synonymous with any combustible
nonpetroleum fuel source which may include coal- and shale-derived fuels
and feedstocks as well as those derived from agricultural products such as
grain, wood, and cellulose. However, industry has become increasingly more
interested in synfuel technologies with products that are easily substi-
tuted, in a marketing and utilization sense, for petroleum and natural gas.
These synfuel technologies are those relating to coal- and shale-derived
products. The following discussion is limited only to these products.

SHALE OIL MAY BE FIRST SYNFUEL TO ENTER THE MARKET AS A PETROLEUM REPLACEMENT

As a direct substitute for large volumes of liquid fuels, oil shale technology is perhaps closest to commercialization in the U.S. Several consortia and companies with established shale oil projects have been engaged in the development of shale oil technology for some time. (reference Table 3). These projects are all located in prime shale areas of Colorado and Utah.

Many technologies are being developed and tested which are aimed at extracting kerogen, a waxy organic material, from shale. Most involve heating shale to about 480°C and pyrolyzing the kerogen into a viscous liquid called shale oil. They differ in the manner in which this heating process is accomplished; surface retorting, in situ retorting, or modified in situ retorting.

In surface retorting, oil shale is mined, crushed to the proper size and then fed to a large kiln for heating. Several surface retorting processes are under development and they differ primarily in the heating method employed. Internal-combustion retorting heats shale by the circulation of hot gases that are produced inside the retort by the combustion of residual carbons in the shale. Gas-cycle retorts used by Union Oil heat the shale by circulating externally heated fluids. No combustion occurs inside the retort. In solid-heat-carrier retorting, shale is mixed with hot solids that are heated outside the retort and cycled through the shale. TOSCO II is an example of this method, using ceramic balls as the heat carrier.

In situ retorting pyrolyzes oil shale while it is still in the ground. The shale bed is ignited and sustained by injection wells, the shale is pyrolyzed, and the oil produced is pumped out of the retort volume through a production well. The spent shale remains in place. For successful in situ retorting, the shale bed must be made permeable to the flow of heat and product oil; various techniques of bed leaching or fracturing are employed. The difficulty of creating a permeable shale bed has led to the development of modified in situ processes. Vertical modified in situ

Table 3. U.S. Oil Shale Projects

Project	Location[1]	Technology	Production Capacity Goal (bbl/day)	Status
Chevron	Piceance Basin	Undecided	50,000	Technical assessment phase.
Colony (TOSCO, EXXON C)	Parachute Creek	Surface retorting	47,000	Construction of commercial modules scheduled for 1980.
Equity Oil	Piceance Creek	Solution injection, modified in-situ	-	Steam-injection feasibility
Geokinetics, Inc.	Uinta County	Horizontal modified in-situ	7-13 2,000-5,000	Several small retorts successfully burned; work on larger retorts in progress.
Getty Oil	Piceance Basin	Surface thermal extraction	-	Getty R&D proposal being considered by DOE
Mobil	Piceance Basin	Undecided	50,000	May start module in 1987
Occidental Oil	Logan Wash	Vertical modified in-situ	70,000	Six retorts burned; 40,000 bbl produced. Retorts 7 and 8 scheduled for cluster burn.
Occidental Oil - Tenneco	Tract C-b, Piceance Basin	Vertical modified in-situ	57,000	Shaft sinking in progress; construction of initial retorts scheduled for 1982.
Paraho (Development Engineering, Inc.)	Anvil Points	Surface retorting	150-200	Shut down due to lack of funding; 88,225 bbl produced over about one year period.
Rio Blanco (Gulf, Amoco)	Tract C-a, Piceance Basin	Vertical modified in-situ, surface retorting	50,000	Modular program consisting of 5 retorts scheduled for completion by 1982.
Superior Oil	Piceance Creek	Multimineral recovery, surface retorting	13,000	Company seeking land exchange with Federal Government which was denied in Febuary 1980.
TOSCO-Sand Wash	Uinta Basin	Modified in-situ, surface retorting	50,000	Feasibility studies in progress
Union Oil	Parachute Creek	Surface retorting	50,000	Construction of experimental mine and plant scheduled for 1982.
White River (Sohio, Sunoco, Phillips)	Tracts U-a and U-b, Uinta Basin	Modified in-situ, surface retorting	100,000	Operations suspended due to legal proceedings on ownership of lands.

[1]State Locations: Piceance Basin - Colorado; Parachute Creek - Colorado; Uinta County - Utah; Logan Wash - Colorado; Anvil Points - Colorado.

(VMIS) retorting removes a portion of the shale from the bottom of the deposit and fractures the remaining shale to create a chimney of shale rubble. The shale is retorted in this chimney from top to bottom. Occidental Oil Company has been testing VMIS retorting on shales at Logan Wash and Piceance Creek Basin in Colorado. Horizontal modified in situ retorting lifts the overburden in some cases, and fractures the shale seam to retort the shale from side to side. Geokinetics, Inc. is developing this technique in Utah.

The technology for surface retorting is more advanced than in situ retorting. Process variables are easier to monitor and control in above ground retorts than in underground retorts. However, large-scale commercial surface retorting requires large-scale oil shale mining, hauling, and crushing; and large-scale disposal of spent shale. It is also limited to that portion of the shale resources that is mineable. In situ retorting without mining is applicable to a greater variety of shale beds, and eliminates the requirements for handling, crushing, and spent shale disposal. Attempts to demonstrate this technology have identified many development problems. Modified in situ processes present a compromise, requiring some mining and handling, but offering more process control and easier development.

The crude shale oil produced by retorting will be upgraded by further processing. This upgraded shale oil, or syncrude, will be used as a refinery feedstock or boiler fuel. It is well suited for refining into middle distillate fuels. If hydrocracking is chosen for the refining process, the yield and range of products is particularly desirable: motor gasoline - 17 percent; jet fuel - 20 percent; diesel fuel - 54 percent; and residuals - 9 percent.

Several oil shale projects, with identified participants, plan to begin operation during the 1980s. The technologies, which are proprietary in many cases, appear to be sufficiently mature to move ahead to commercialization. Several retorts have been successfully operated by Geokinetics, Inc., Occidental Oil, Paraho, Union, and TOSCO. Colony, Union Oil, and Occidental Oil have announced plans to begin commercial development in 1980. All technologies have been demonstrated at pilot scale or larger.

BOTH DIRECT AND INDIRECT ROUTES TO COAL LIQUIDS ARE AVAILABLE

DEMONSTRATION AND FULL-SCALE UNITS ARE BEING ENGINEERED

Coal, hydrogen, and a coal-derived oil are mixed at high temperature and pressure to accomplish direct liquefaction. Under these conditions, the coal decomposes, and the fragments react with hydrogen to form additional derived oil, which is separated from the unreacted solids and further refined to produce usable liquid fuels. Indirect liquefaction processes react the coal with oxygen and steam in a gasifier to produce a synthesis gas composed mainly of carbon monoxide, carbon dioxide, and hydrogen. After the carbon dioxide and other impurities are removed from the gas, the carbon monoxide and hydrogen are chemically combined in a catalytic reactor to produce liquid products for use as chemical feedstocks or liquid fuels.

There are three major direct coal liquefaction processes currently undergoing development: SRC II, Exxon Donor Solvent (EDS), and H-Coal (reference Table 4). These processes differ mainly in the manner in which the hydrogen is made to react with coal fragments to produce the unrefined coal liquids. In the SRC II process, the coal feed and hydrogen are mixed with a process recycle stream that contains unreacted coal ash as well as coal-derived oil. The iron pyrite in the unreacted ash catalyzes the reaction between the coal fragments and hydrogen. In the EDS process, the coal feed and hydrogen are mixed with a specially hydrogenated coal oil called the donor solvent. The hydrogen added to the coal fragments is provided by the solvent and the hydrogen gas mixed in the reactor. The donor solvent is made by catalytically hydrogenating coal-derived oil using conventional petroleum refinery hydrotreating technology. In the H-Coal process, the unreacted coal and hydrogen are mixed with coal-derived oil and an added solid catalyst in a special reactor referred to as an ebullated bed.

Once the gases and distillable liquid products have been separated from the reactor effluent, the remaining "bottoms" material is processed. This material contains significant quantities of heavy hydrocarbons which must be efficiently utilized to enhance process economics. The principal bottoms processing step under consideration for the EDS process is

Table 4. Major Coal Liquefaction Processes

PROCESS	PROCESS TYPE	PRODUCTS	STATUS
Solvent Refined Coal, SRC II (Gulf Oil)	Direct liquefaction by solvent extraction: coal dissolved in solvent, slurry recycled, catalytic hydrogenation	LPG Naphtha Fuel Oil SNG	Pilot Plant under operation. 6700 ton/day of coal (20,000 barrels/day of oil equivalent) demonstration module under design and schedule for operation in 1984-1985
H-Coal (Hydrocarbon Research, Inc.)	Direct liquefaction by catalytic hydrogenation, ebullated catalyst bed	Naphtha Fuel Oil	600 ton/day (1400 barrels/day of oil equivalent) pilot plant under construction, testing will begin in 1980. Plant is located at Catlettsburg, Kentucky
Exxon Donor Solvent, EDS (Exxon Research and Engineering Company)	Direct liquefaction by extraction and catalytic hydrogenation of recycled donor solvent	Propane Butane Butane Naphtha Fuel Oil	250 ton/day (500 barrels/day of oil equivalent) pilot plant under construction, testing will begin in 1980. Plant is located at Baytown, Texas
Fischer-Tropsch (M.W. Kellogg/Lurgi)	Indirect liquefaction, liquefaction of synthesis gas in an fluid bed catalytic converter	Gasoline LPG Diesel Fuel Heavy Fuel Oil Medium Btu Gas SNG	SASOL I, 800 tons/day, producing over 10,000 bbl day of liquids in commercial production since 1956. SASOL II, 40,000 tons/day, producing over 50,000 bbl day of liquids has been completed and will begin start-up in 1980. SASOL III with approximately the same capacity as SASOL II is currently being planned.
Mobil M	Indirect liquefaction, liquefaction of synthesis gas in fixed bed using molecular size-specific zeolite catalyst	Gasoline LPG	Commercial scale plant to produce 12,500 barrels of gasoline using reformed natural gas is planned for New Zealand in 1984-1985

FLEXICOKING, which consists of thermal cracking of the bottoms to produce additional liquids and coke. The coke is subsequently gasified to produce plant fuel gas or hydrogen for the liquefaction step. Bottoms processing for the SRC II and H-Coal processes probably will be partial oxidation (i.e., gasification) to produce hydrogen for the liquefaction step.

There are two major indirect coal liquefaction processes: Fischer-Tropsch which is commercial now in South Africa, and Mobil-M which is expected to be commercial in 1983-84. In the Fischer-Tropsch process, the purified synthesis gas from the gasifier is reacted over an iron catalyst to produce a broad range of products extending from lightweight gases to heavy fuel oil. The broad product distribution from this process is generally considered as a disadvantage where large yields of gasoline are desired. Improved catalysts are currently being developed at the bench scale to maximize the yield of gasoline-range hydrocarbons. In the Mobil-M process, the synthesis gas is first converted to methanol using commercially available technology. The methanol is then catalytically converted to high-octane gasoline over a molecular-size-specific zeolite catalyst.

Indirect coal liquefaction is successfully operating on a commercial scale at the SASOL I plant in South Africa using the Fischer-Tropsch technology. The SASOL I plant produces gasoline, jet fuel, diesel oil, middle distillates, and heavy oil. SASOL II, producing 50,000 barrels per day of coal-derived liquids, has been completed and will begin operation later in 1980. Active interest in this technology has developed and plans to license and construct similar plants in the U.S. are progressing. There is strong interest in the Mobil-M gasoline indirect process because of its attractive high-octane gasoline yield. A commercial-scale plant producing 12,500 barrels per day of gasoline is planned for operation in New Zealand by 1985.

Direct coal liquefaction technologies are in various stages of development. SRC I and II processes have been tested at the pilot plant level and are entering into the demonstration plant stage.

Large pilot plants are currently under construction for testing of the H-Coal and EDS processes. These plants are located at Catlettsburg, Kentucky, and Baytown, Texas, respectively.

SRC I process produces primarily a solid product with a small amount of useful liquid product. However, SRC II process produces primarily liquid products.

In addition to these major coal liquefaction technologies, several other processes have received attention, including the Dow process, Riser Cracking, Synthoil, and the Zinc Halide process. All have been tested in small-scale units.

A WIDE VARIETY OF COAL MAY BE USED IN THE SYNFUELS INDUSTRY

Most coal gasifiers react coal, steam, and oxygen to produce a gas containing carbon monoxide, carbon dioxide, and hydrogen. When air is used as the oxygen source, the product gas contains up to 50 percent nitrogen and is referred to as low Btu gas since its heat of combustion is only 80 to 150 Btu/standard cubic feet (scf). Synthesis gas or medium-Btu gas ranges from 300 to 500 Btu/scf.

Low-Btu gas is used as a fuel gas near its point of generation since its low heating value makes it uneconomical to distribute over long distances. Medium-Btu gas can be used as a fuel gas and transported economically over distances of up to 200 miles. It can also be used as a chemical feedstock for the production of methanol or gasoline. Finally, it can be converted catalytically to substitute natural gas (SNG), having a heating value of about 1,000 Btu/scf. Additionally, medium-Btu gasification is an integral part of all indirect liquefaction technologies.

There are many coal gasification technologies that differ in design and operation, depending upon the type of coal used and the product desired. High- and medium-Btu gasification technologies using noncaking coals characteristic of U.S. western coals are relatively well developed. Severe operational problems are encountered with commercially available gasifiers in processing caking coal such as those found in the eastern U.S. Several gasification technologies for high- and medium-Btu gases are under active development (reference Table 5). Many additional processes are being tested, but at less advanced stages of development (reference Table 6).

A fixed-bed gasifier, such as the Lurgi, feeds coal to the top of the gasifier. The descending coal is successively dried, devolatilized, and gasified in contact with gases rising from the bottom. Steam and oxygen are introduced at the bottom of the gasifier, and solid ash is removed through an ash lock. In some gasifiers, such as British Gas Company (BGC) Lurgi, the temperature at the bottom of the bed is sufficient to melt the

Table 5. Coal Gasifiers for High, Medium and Low Btu Gas

Process	Process Type	Potential Products	Most Suitable Products	Status
Lurgi Dry Ash	Pressurized fixed bed, dry bottom	Substitute Natural Gas (SNG, also known as High Btu Gas), Medium Btu Fuel[a] Gas, Low Btu Fuel Gas	SNG, Medium Btu Fuel Gas, Low Btu Fuel Gas	40 years of commercial development and 14 commercial plants located in Australia, Germany, UK, India, Pakistan, South Africa, Korea. Average module size 800 tons/day (2000 BOE)[c]
British Gas Company (BGC) Lurgi	Pressurized fixed bed slagging bottom	SNG, Medium Btu Fuel Gas, Low Btu Fuel Gas	SNG, Medium Btu Fuel Gas, Low Btu Fuel Gas	790 tons/day (of coal) (2000 BOE) pilot plant tested in Westfield, Scotland
Texaco	Pressurized single stage entrained, slurry feed	SNG, Medium Btu Synthesis Gas, Low Btu Fuel Gas	Medium Btu Synthesis[b] Gas	160 ton/day (400 BOE) plant operating in West Germany
U-Gas Institute of Gas Technology (IGT)	Pressurized fluid bed, ash agglomerating	SNG, Medium Btu Fuel Gas, Low Btu Fuel Gas	Medium Btu Fuel Gas	14000 tons/day of coal plant (35000 BOE) producing Medium Btu Fuel Gas, under design for construction in Tennessee
Westinghouse	Pressurized single stage fluid bed, ash agglomerating	SNG, Medium Btu Fuel Gas, Low Btu Fuel Gas	SNG, Medium Btu Fuel Gas	15 ton/day (40 BOE) process development unit, under testing at Waltz Mill, Pa.
Shell Koppers	Pressurized entrained, dry feed	Medium Btu Synthesis Gas, Low Btu Fuel Gas	Medium Btu Synthesis Gas	150 ton/day (400 BOE) pilot plant in operation in W. Germany. 1,000 ton/day scheduled in 1983/1984.
Koppers-Totzek	Atmospheric entrained, dry feed	Medium Btu Synthesis Gas, Low Btu Fuel Gas	Medium Btu Synthesis Gas	1,000 ton/day (2500 BOE) plant in operation in South Africa for the production of ammonia

a Medium Btu Gas with significant concentration of methane is more suitable for use as fuel, and therefore identified as Medium Btu Fuel Gas.

b Medium Btu Gas with low concentration of methane is more suitable for chemical synthesis, and therefore identified as Medium Btu Synthesis Gas

c BOE - Barrels per day of oil equivalent

Table 6. Status of Other Coal Gasification Processes

DEMONSTRATION PLANTS	SCALE (tons/day coal feed)	STATUS
HYGAS	7340	Conceptual Design[a]
COED-COGAS	2210	Detailed Design[b]
U-GAS	3160	Detailed Design

PILOT PLANTS/PDUs

BELL HIGH MASS FLUX	6	Operational[c]
BIGAS	120	Operational
COMBUSTION ENGINEERING	5	Operational
DOW	24	Under Construction
EXXON CATALYTIC	100	Proposed
GEGAS	24	Operational
HYDRANE	4	Proposed
MOLTEN SALT	24	Operational
MOUNTAIN FUEL	12	Proposed
SYNTHANE	72	Mothballed
TRI-GAS	1	Operational

---

[a]Conceptual design incorporates all important details of major unit areas in the plant. Material balances are provided around all major unit areas. (Unit area is a section of the plant consisting of several components integrated to perform a single transformation on the product stream. Examples are gasification, raw gas cooling, gas cleanup, or methanation.)

[b]All equipment and detailed pipeline diagrams are prepared as part of detailed design. In addition, detailed material balances are prepared for each piece of equipment.

[c]The plant is either operating or has operated successfully in the past.

ash, allowing its removal as molten slag. The slagging feature provides a distinct advantage in contending with the caking characteristics of eastern U.S. coals.

Lurgi high-pressure operation, in conjunction with relatively low gasification temperatures, favors the formation of significant quantities of methane in the gasifier, enhancing the heating value of the product. These conditions also favor production of by-products such as tars and impurities like phenols, organic nitrogen compounds, and sulfur compounds.

In fluid-bed gasifiers currently under development, high-velocity gases pass up through the bed to fluidize the coal, providing excellent mixing and temperature uniformity throughout the reactor. Operability with caking coals (eastern U.S.), as well as low tar production and tolerance to upsets in fuel rates, has been demonstrated at the pilot scale for both the Westinghouse and U-Gas gasifiers.

The Texaco and Koppers-Totzek gasifiers are representative of entrained-bed technology in which the solid particles are concurrently entrained in the gaseous flow. Flame temperatures at the burner discharges are in the range of 1370 to 1925°C, resulting in melting of the coal ash with minimum production of impurities. Entrained-flow gasifiers may be favored for the production of synthesis gas for indirect liquefaction. They can operate with caking coals. However, compared to fluid-bed gasifiers, they have very low carbon holdup capability in the reactor and, therefore, have limited safeguard against possible formation of explosive mixture in the reactor in case of coal feed interruption.

There has been extensive commercial experience in the U.S. with low-Btu coal gasification technologies operating near atmospheric pressure. However, these applications have been limited to small-scale captive applications for providing industrial process heat and space heating. For example, the Wellman-Galusha gasifier designed for atmospheric pressure operation was used extensively by industry years before pipeline-supplied natural gas was readily available at comparatively lower cost. Pressurized gasification processes capable of yielding high-Btu gas for pipeline use and medium-Btu gas for chemical feedstocks are less developed, with the exception of the Lurgi fixed-bed process. The Lurgi process is based on 40 years of commercial development at 14 commercial plants that are located in

Australia, Germany, U.K., Korea, India, Pakistan, and South Africa. A great deal of interest in the Lurgi technology is emerging in the U.S. with several announced plans for SNG production by pipeline and gas utility companies. Several projects utilizing the Texaco process for captive applications (chemical feedstocks and on-site power generation) are in the planning and design stage with at least one project (Tennessee-Eastman) scheduled for construction in 1980.

DEVELOPMENT OF THE SYNFUELS INDUSTRY OVER THE NEXT 20 YEARS

Three scenarios or projections of synfuel industry buildup rates to the year 2000 have been developed to illustrate the potential range of synfuel product utilization:

- A "National Goal" scenario driven by Federal incentives

- A "nominal production" or most likely scenario

- An "accelerated production" scenario representing an upper bound for industry buildup.

ACHIEVING THE NATIONAL GOAL - SCENARIO I

In July 1979, President Carter announced new energy initiatives for the U.S. aimed at reducing our dependence on imported oil. One of the key elements of this policy is the provision of Federal funds to stimulate production of synthetic fuels at the rate of 2.2 million barrels per day (MMBPD) by 1992. Specifically, the national synfuel goals are:

Coal Liquids. To stimulate and accelerate the construction and operation of the first few plants to provide sufficient data on the competing commercial coal liquefaction processes so that industry, with its own investment, stimulated by Government incentives if required, will build plants with sufficient capacity to provide upwards to 1 MMBPD liquid fuels by the year 1992.

Shale oil. To stimulate shale oil production at the rate of 0.4 MMBPD by 1990.

High-Btu Gas. To develop and implement a program that enables the U.S., by 1992, to produce significant quantities of pipeline quality gas (0.5 MMBPD - oil equivalent[*]) from commercial HBG plants in an environmentally acceptable manner. This is facilitated by the short-range goal of having two or three commercial HBG plants in operation by the mid-1980s.

---

[*]For easy comparison with petroleum supply/demand figures, synfuel production rates are expressed in barrels of oil equivalent in this document. This does not imply that high-, medium- and low-Btu gases from coal that are substituted for domestic natural gas will have any direct effect on the reduction of imported oil.

Low-/Medium-Btu Gas. To stimulate an initial near-term commercial capability for several medium-Btu commercial plants in key industries as well as utilities, for energy and feedstock applications for both single and multiplant use, and for multiple applications of low-Btu gas in each of the prime industry markets. Commercial-scale development will depend on the long-term economics of this technology, vis-a-vis the price of domestic oil and natural gas. Once a capability has been established, capacity will be accelerated to achieve at least 0.29 MMBPD oil equivalent by 1992. Of this total, up to 0.04 MMBPD oil equivalent will be provided from 40 to 50 low-Btu facilities and up to 0.25 MMBPD oil equivalent from 25 to 30 medium-Btu plants. Again, it must be mentioned, that if this low- and medium-Btu coal-gas is substituted for natural gas, there will not be a direct effect on the reduction of imported oil.

The key assumptions allowing achievement of these goals are: (1) Federal funds provided are sufficient to reduce investment risk by the synfuel industry through 1992, and (2) other requirements for industry development are satisfied, i.e., environmental permits, material, equipment, and labor. A likely buildup rate profile for the synfuel industries under this scenario is shown in Figure 1.
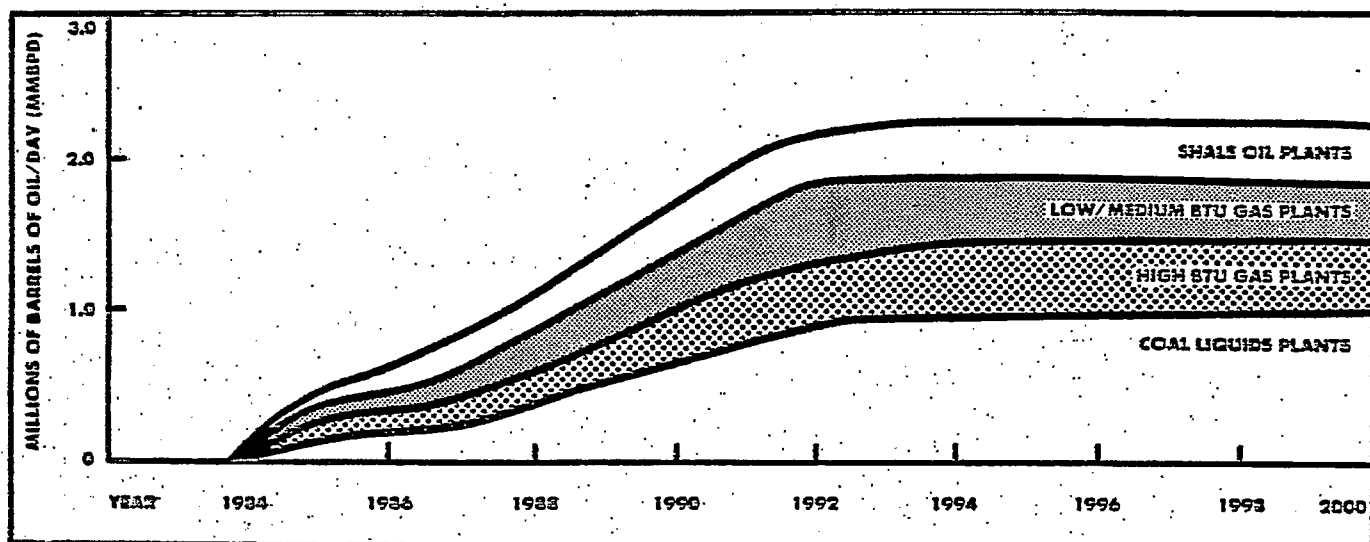

Figure 1. Synfuels Industry Buildup for the National Goal Scenario

227

For shale oil, several of the most advanced projects were selected as a basis. The planned operation startup schedules and capacity buildup rates for these projects were used to generate the industry production buildup profile to about 0.4 MMBPD by 1992. The period beyond 1992 is viewed as one of technology consolidation: gaining a firm footing with regard to environmental and economic performance and technology improvements. This type of industry production profile is not without precedent; for example, the Federal support of the synthetic rubber industry during World War II.

The goal of 1 MMBPD of coal liquids will be met predominantly by indirect coal liquefaction. At present, the only commercially demonstrated coal liquefaction process is the Fischer-Tropsch embodied in the SASOL plants in South Africa. The Mobil-M process should be commercially demonstrated within the next five years. Considering construction and permitting lead times, plants of this type could begin operation around 1985. To meet the production goal, 10 to 15 plants of a nominal 0.05 MMBPD capacity must be in operation by 1992. A potential drawback to the commercialization of SASOL technology in the U.S. is the broad product distribution, ranging from light hydrocarbon gases to heavy fuel oil. The Mobil-M technology, on the other hand, produces an all-gasoline product which would be particularly well suited to the U.S. market demands. Given this apparent advantage of Mobil-M technology over SASOL, it is believed that industry should favor commercialization of both Mobil-M and SASOL technology during the next few years, with the breakdown being roughly 50/50. Approximately 75 percent of the coal liquids production will be due to these indirect liquefaction processes.

For the direct liquefaction processes, there will not be sufficient experience and information to attract any more than developmental interest over the next few years, under this scenario. By 1985 there should be sufficient information available from the operation of the EDS, H-Coal and SRC II plants to support a commercialization decision concerning these processes. Federal incentives will likely be distributed such that by 1992, three or four pioneer commercial-scale plants employing direct liquefaction will begin to appear. Of the total production goal of 1 MMBPD of coal liquids it is estimated that 25 percent will be produced by these

first commercial direct liquefaction plants embodying the basic SRC II, EDS and H-Coal technologies, or improvements and modifications to these. It is projected that for the next few years after 1992, production will remain at 1 MMBPD while technological evaluations are performed. These direct lique-faction plants will be located near the major eastern U.S. coal areas.

The Lurgi fixed-bed process is the lead high-Btu coal gasification technology and has been commercially demonstrated outside the U.S. It is expected to be utilized in all commercial plants constructed over the next 10 years. As the process requires noncaking coals, these plants will most likely be located in the western U.S. Interest will continue in other high-Btu gasification technologies such as the Slagging Lurgi which is capable of using eastern caking coals. At least one of these alternate or advanced processes probably will be supported under Federal incentives but it is unlikely that a commercial plant will appear until the early 1990s, and this would probably be located near a midwestern coal resource.

The Lurgi fixed-bed medium-Btu process is the lead technology for medium-Btu gas. Texaco partial oxidation gasification or similar pressur-ized entrained-bed gasifiers such as pressurized Koppers-Totzek, will be under development and demonstration during the early 1980s and will likely serve as the prime medium-Btu gasification process for eastern coals. To 1992, however, the major buildup in medium-Btu gasification will come from Lurgi plants located in the western U.S.

For low-Btu gasification, the several technologies that are currently available and providing commercial service are assumed to be easily applied, under the incentives existing to 1992, to generate the 0.04 MMBPD production rate goal. Low-Btu gas will generally be captively employed as fuel gas or used on-site for combined-cycle power generation.

The production buildup profile for major synfuel products resulting from of the synfuels industry buildup in Scenario I is shown in Figure 2. These product quantities are projected to enter commercial use and are to be considered in assessments of potential environmental impacts from synfuels. Naturally, these major products are presented for the sake of clarity, but there are many other products and byproducts that will be produced and distributed into the market place. These products and byproducts will also vary in greater or lesser quantities in Scenarios II and III which follow.   229
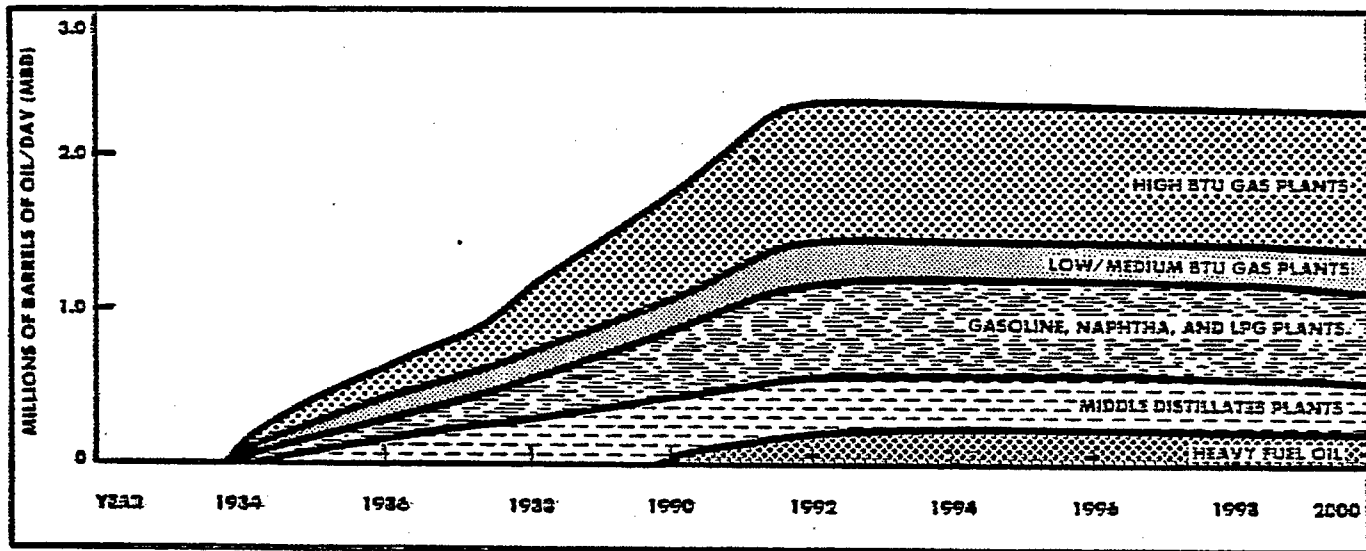
Figure 2. Major Synfuel Product Buildup for
the National Goal Scenario

PRODUCTION AT A NOMINAL RATE - SCENARIO II

Recent studies of the technical capability of the U.S. to meet the synfuel national goal point out that there are significant concerns regarding achieving this goal. They include:

- Availability of skilled manpower: it is expected that the supply of engineers and construction labor will be severely taxed to meet the synfuel production goal set forth in Scenario I.

- Availability of critical equipment: certain critical equipment for the synfuel industry such as compressors, heat exchangers, and pressure vessels are expected to be in short supply unless corrective measures are taken now, thus slowing the synfuel industry buildup rate indicated in Scenario I.

- Diversion of investment to competing technologies: demand on the limited capital available in the economy by competing energy supply technologies, such as coal liquefaction, coal gasification, oil shale, geothermal, and solar technologies, could result in the slowing of buildup rates for some technologies.

- Environmental data: lack of environmental data needed for regulatory approvals could slow down the buildup rate.

230

• Licensing: time and construction schedule constraints imposed by State and Federal licensing and permitting requirements could hinder synfuel industry buildup rate.

Taking these concerns into consideration, a nominal synfuels production buildup - Scenario II - has been developed, as indicated in Figure 3. A production rate of about 2.1 MMBPD is estimated by the year 2000, instead of 1990 as indicated in Scenario I. The technologies expected to contribute to both Scenarios I and II are the same; the major difference is in the rate of buildup: it is slower and delayed in time.
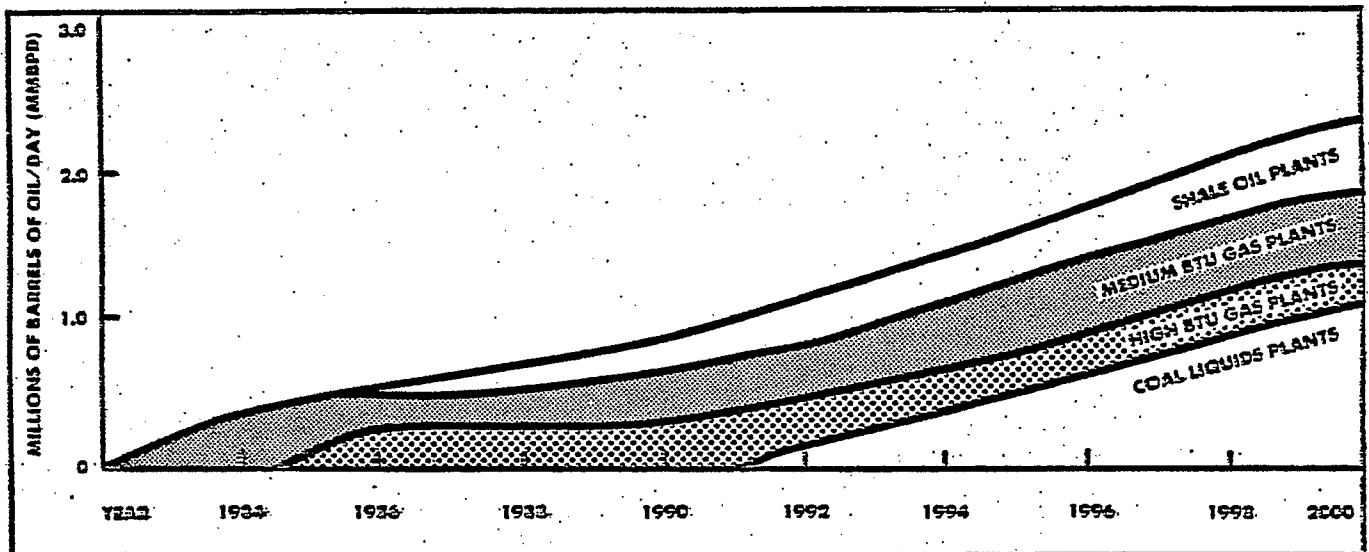
Figure 3. Synfuels Industry Buildup for the Nominal Production Scenario

For shale oil, a nominal production rate of 0.4 MMBPD should be achieved by the year 2000. The buildup rate is estimated to lag about 4 years behind that of Scenario I and is based on the following observations:

● Some technologies are still considered developmental, such as the modified in situ process.

● Land problems, including availability of off-tract disposal sites, may take longer to resolve.

Under this scenario, no large-scale commercial coal liquefaction plants are projected to be on line until 1992 with a growth rate beyond yielding 1 MMBPD by the year 2000. It is believed that Federal incentives will be applied to support construction of one each of the indirect

231

liquefaction plants and a direct liquefaction plant only after sufficient assessment has been made of the operations of the EDS and H-Coal pilot plants and the SRC II demonstration plant. Rather than commit sizable resources to the commercialization of indirect liquefaction, a decision probably will be delayed resulting in no operating commercial liquefaction plants before 1992 under this scenario. During the 1980s it is believed that improvements will be made in both the operating indirect liquefaction plants and the designs of the direct liquefaction processes. These "advanced" technologies with product slates yielding primarily transportation fuels, will be sufficiently attractive to encourage development of 1 MMBPD of coal-derived liquid production by the year 2000.

Currently there is a great deal of interest in SNG technology. Several gas utility and pipeline companies have expressed plans to construct high-Btu plants. With incentives, several of these plants will be constructed and in operation by 1985. However, as a result of the projected improved outlook for gas supplies, including potential from unconventional sources, the availability of "imported" conventional natural gas (Alaskan, Canadian and Mexican) and the current unfavorable rate-structure pricing policy, the complete commercialization of HBG will be hampered. Its production rate is not likely to expand beyond the 0.25 MMBPD-level attained around 1992 under this scenario.

The buildup of medium-Btu gas plants will also be impeded by the availability of natural gas; however, for certain industrial applications requiring large volumes of uninterrupted supplies (e.g., chemical feedstocks, cogeneration) low-/medium-Btu plants will remain attractive. It is estimated that production of low-/medium-Btu gas will reach a level of 0.45 MMBPD by 1992.

ACCELERATED PRODUCTION - SCENARIO III

The accelerated production scenario is based on the assumption that Federal incentives are sufficient to synfuels production to meet the national goals in 1992, that operation of synfuels plants up to 1992 is successful to the extent confidence in processes is gained, and all resource requirements are satisfied. Licensing and permitting procedures must also be streamlined. It is assumed that demand for coal-derived synfuels remains at a level such that new plant capacity continues to be

added to the year 2000 at about the same rate as the buildup to 1992. For shale oil, the production of 0.9 MMBPD by the year 2000 is based on a survey and analysis of the desired goals of each industrial developer. As indicated in Figure 4, a total synfuels production rate of 5 MMBPD may be reached by the year 2000. This includes 2.6 MMBPD of coal liquids, 1.5 MMBPD of gas and 0.9 MMBPD of shale oil.
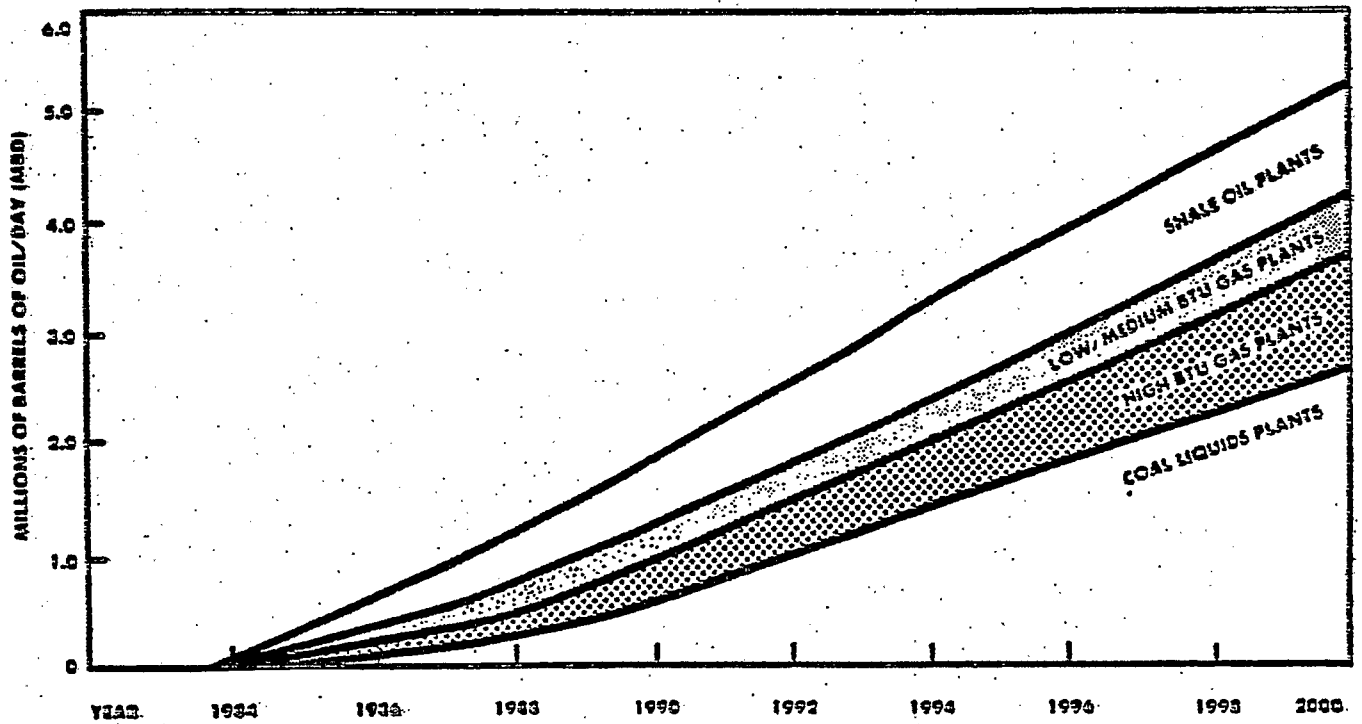
Figure 4. Synfuels Industry Buildup for the
Accelerated Production Scenario

However, in view of the limitations facing the synfuel industry, some of which were discussed earlier, the accelerated production scenario is highly unlikely. The synfuels industry buildup rate (Figure 4) for this scenario can be considered an upper bound to synfuels utilization over the next 20 years.

The three scenarios describing possible synfuel industry buildup profiles provide a basis for projecting the market penetration of synfuel products in the near future. As these products enter the market, potential environmental impacts related to synfuels utilization must be considered.

THERE IS A LARGE POTENTIAL MARKET FOR SYNFUEL PRODUCTS AND BY-PRODUCTS

The major synfuel products could be broadly classified into five groups:

- Gaseous Products

  - High-Btu gas
  - Medium-Btu gas
  - Low-Btu gas
  - Liquified Petroleum Gas (LPG)

- Light Distillates

  - Gasoline
  - Naphtha

- Middle Distillates

  - Jet fuel
  - Kerosenes
  - Diesel oil

- Residue

  - Heavy fuel oil
  - Lubricants

- Petrochemicals.

GASEOUS PRODUCTS

The high- and medium-Btu gases are suitable for essentially all industrial fuel applications that can be serviced by coal, oil or natural gas. In some cases equipment modifications or special controls will have to be implemented to retrofit existing plants for medium-Btu gas, whereas this problem may not exist for high-Btu gas installation. However, there should be no difficulty in employing either high- or medium-Btu gas in new industrial installations. These products will be utilized by major energy consuming industries such as food, textile, pulp and paper, chemicals, and steel. It appears that only chemical, petroleum, and steel industries will require sufficient fuel gas at a single location to economically justify the dedication of a single gasification plant. Other industrial plants will have to share the output distributed by pipeline from a central gasifier, or tap into the existing natural gas pipeline system for their need. Preliminary economic studies indicate that it is not economical to

transport medium Btu gas through pipelines for more than 200 miles. Medium-Btu gas can also be utilized by the petrochemical industries as chemical feedstock for the production of ammonia, methanol, and formaldehyde. Currently most of this requirement is met by reforming natural gas. The use of medium-Btu gasification appears especially attractive when integrated with new combined plants for utility applications.

The major characteristics of low-Btu gas are its high nitrogen content, low carbonmonoxide and hydrogen content, and resulting heating value typically below 150 Btu/SCF. Its flame temperature is also about 13 percent lower than that of natural gas. Because of these characteristics low-Btu gas is limited to on-site use, industrial processes requiring temperature below 2800°-$3000^{\circ}F$, and is generally unsuitable for use as a chemical feedstock. Further, because of its low energy density it requires significant equipment modifications for retrofit applications. Today there are operating and planned low-Btu gasifiers in the U.S. for:

- Kiln firing of bricks
- Iron ore pelletizing
- Chemical furnace
- Small boilers

Liquified petroleum gas (LPG) has applications for industrial, domestic, and transportation uses. In domestic applications LPG is used mainly as a fuel for cooking and for water and space heating. In industry, LPG finds a large number of diverse outlets. Apart from use as a fuel in processes which require careful temperature control (glass and ceramics, electronics) or clean combustion gases (drying of milk, coffee, etc.), LPG is also used in the metallurgical industry to produce protective atmospheres for metal cutting and other uses. The chemical industry, particularly on the U.S. Gulf Coast, uses petroleum gases for cracking to ethylene and propylene as well as for the manufacture of synthesis gas. Small portions of LPG are also used to fuel automotive vehicles. Another use of LPG is to enrich lean gas made from other raw materials to establish proper heating value levels. On a volume basis, production of LPG in the U.S. exceeds that of kerosene and approaches that of diesel fuel. About 40 percent of LPG production is used by the chemical industry, another 40 percent is for domestic use, 10 percent for automotive use, and the

236

remaining distributed among other industrial and agricultural fuel uses. Currently LPG is supplied primarily from refineries handling petroleum crudes. With the anticipated shortfall in the supply of these crudes, the resulting shortage of LPG will be met to some extent by LPG from synfuel plants.

LIGHT DISTILLATES

Gasoline, which is a major light distillate, is generally defined as a fuel designed for use in reciprocating, spark ignition internal-combustion engines. Other uses for gasoline are of small volume. Primarily it is used as fuel for automotive ground vehicles of all types, reciprocating aircraft engines, marine engines, tractors and lawn mowers. Other small-scale uses include fuel in appliances such as field stoves, heating and lighting units, and blow torches. By far the primary use of gasoline produced from coal will be for transportation applications. Currently we consume nearly 6.8 MMBPD of petroleum-derived gasoline and this corresponds to about 40 percent of the total petroleum consumption.

Naphthas have a wide variety of properties and serve many industrial and domestic uses. Their primary market is the petrochemical industry where they can be used for the manufacture of solvents, varnish, turpentines, rust-proofing compounds, pharmaceuticals, pesticides, herbicides, and fungicides. However, preliminary analysis indicates that there will be a relatively small amount of coal-derived naphthas entering the market.

MIDDLE DISTILLATES

The market for middle distillates, which essentially are jet fuel, kerosene, diesel oil and light fuel oil, are jet aircraft, gas turbines, and diesel engines used for transportation and stationary applications, and residential and commercial heating.

RESIDUES

The market for residues, consisting mainly of fuel oil, is primarily for industrial, utility and marine fuel use. Other applications for residues include preparation of industrial and automotive lubricants, metallurgical oils, roof coatings, and wood preservative oils. Coke is another likely useful product from residue.

PETROCHEMICALS

Many synfuel products, in addition to their primary use as fuel, are likely to be used by the petrochemical industry for the production of several other by-products. Currently over 3000 petrochemical by-products are derived from petroleum and natural gas sources. These include items like synthetic rubbers, plastics, synthetic fibers, detergents, solvents, sulfur, ammonia and ammonia fertilizers and carbon black.

Petrochemicals from synfuels will generally fall under three broad groups based on their chemical composition and structure: aliphatic, aromatic, and inorganic. An aliphatic petrochemical is an organic compound which has an open chain of carbon atoms. Important petrochemicals in this group include acetic acid, acetic anhydride, acetone, ethyl alcohol, and methyl alcohol. Most aliphatic petrochemicals are currently made from methane, ethane, propane or butane. Aliphatics currently represent over 60 percent of all petrochemicals and are the most important group economically.

An aromatic petrochemical is also an organic compound but one that contains or is derived from a basic benzene ring. Important in this group are benzene, toluene, and xylene, commonly known as the B-T-X group. Benzene is widely used in reactions with other petrochemicals. With ethylene it gives ethyl benzene which is converted to styrene, an important synthetic-rubber component. As a raw material it can be used to make phenol. Another use is in the manufacture of adipic acid for nylon. Toluene is largely used as a solvent in the manufacture of trinitrotoluene for explosives. Xylene is used as a source of material for polyester fibers, isophthalic acid, among other petrochemicals.

An inorganic petrochemical is one which does not contain carbon atoms. Typical here are sulfur, ammonia and its derivatives such as nitric acid, ammonium nitrate, ammonium sulfate.

The different end-use applications of major synfuels products are summarized in Table 7. We see from this discussion that coal-derived synfuel products are likely to be used not only as a fuel, but also in the manufacture of a number of other by-products which will be used in multitudes of other applications.

238

Table 7. Major End-Use Applications of Synfuel Products

Major Synfuel Products	Likely Major End Use Applications
High and medium Btu gas	Food, textile, pulp and paper, chemicals, iron and steel industries; residential/ commercial heating
Low Btu gas	Small boilers, kilns, pelletizing
LPG	Glass, electronics, chemical industries; domestic cooking and heating; automotive
Gasoline	Transportation
Naphtha	Petrochemical industry; solvents; varnish; turpentines
Middle distillates (kerosene, diesel, light fuel oil)	Transportation, gas turbines, residential and commercial heating
Residues	Industrial, utility and marine fuel; matallurgical oils; roof coatings; wood preservatives, lubricants

ANTICIPATED SYNFUELS MARKET PENETRATION IN THE VARIOUS
SECTORS OF THE U.S. ECONOMY WILL EXPAND OVER TIME

As an indication of the time frame over which the EPA must consider issues regarding the use of various synfuel products, market development and penetration of these products must be anticipated. For example, the synfuels market may develop as illustrated in Figures 5, 6 and 7, over 1985-1987, 1988-1990 and 1991-2000 time frames.

SYNFUEL PRODUCT UTILIZATION EMPHASIS, 1985-1987

Oil shale-derived synfuels will be introduced into the petroleum product markets about 1985, and based on Scenario I as much as 0.2 MMBPD of shale oil can enter the market by 1987. The first stage of synfuels market infrastructure development will be oriented towards transportation fuels (reference Figure 5) because oil shale that is hydrotreated can be refined in existing refineries to such products as gasoline, jet fuel, diesel and marine fuels. The bulk of this supply will be in the form of middle distillates comprised of jet fuel and diesel oil. The demand for transportation during the late 1980s is expected to be around 10 MMBPD. Of this, about 5 percent is likely to be consumed by the military sector. It is conceivable, therefore, that the bulk of the shale oil products could be utilized by the military, possibly with a Government synfuel purchase guarantee program.

It is anticipated that the oil shale industry will continue to grow producing as much as 0.45 MMBPD by year 2000 as per Scenario I and II and as much as 0.9 MMBPD as per Scenario III. The Bulk of this production is anticipated for the transportation sector.

SYNFUEL PRODUCT UTILIZATION ADDITIONS, 1988-1990

Subsequent buildup of the synfuels industry during the 1988-1990 time period (reference Figure 6) is expected to come from commercial-size, high-Btu gasifiers. As per Scenario 1, the output from these high-Btu gasifiers may be as high as 0.4 MMBPD of oil equivalent by 1990; however, the conservative estimate based on Scenario II is that only around 0.17 MMBPD of oil equivalent is likely to be produced by that time. The high-Btu gasification will serve some of the energy needs of both the industrial and residential/commercial sectors as direct gas sales or through electric

SYNFUEL UTILIZATION DURING 1985-1987

MAJOR SYNFUEL PRODUCT(S)/BYPRODUCTS

Oil Shale Products

- Gasoline
- Middle Distillates
  (e.g., Jet Fuel, Diesel, Kerosene, Light Fuel Oil)
- Residuals
  (e.g., Marine Fuels, Lubricants)

UTILITIES

TRANSPORTATION SECTOR

INDUSTRIAL SECTOR

COMMERCIAL & RESIDENTIAL SECTOR

OIL SHALE
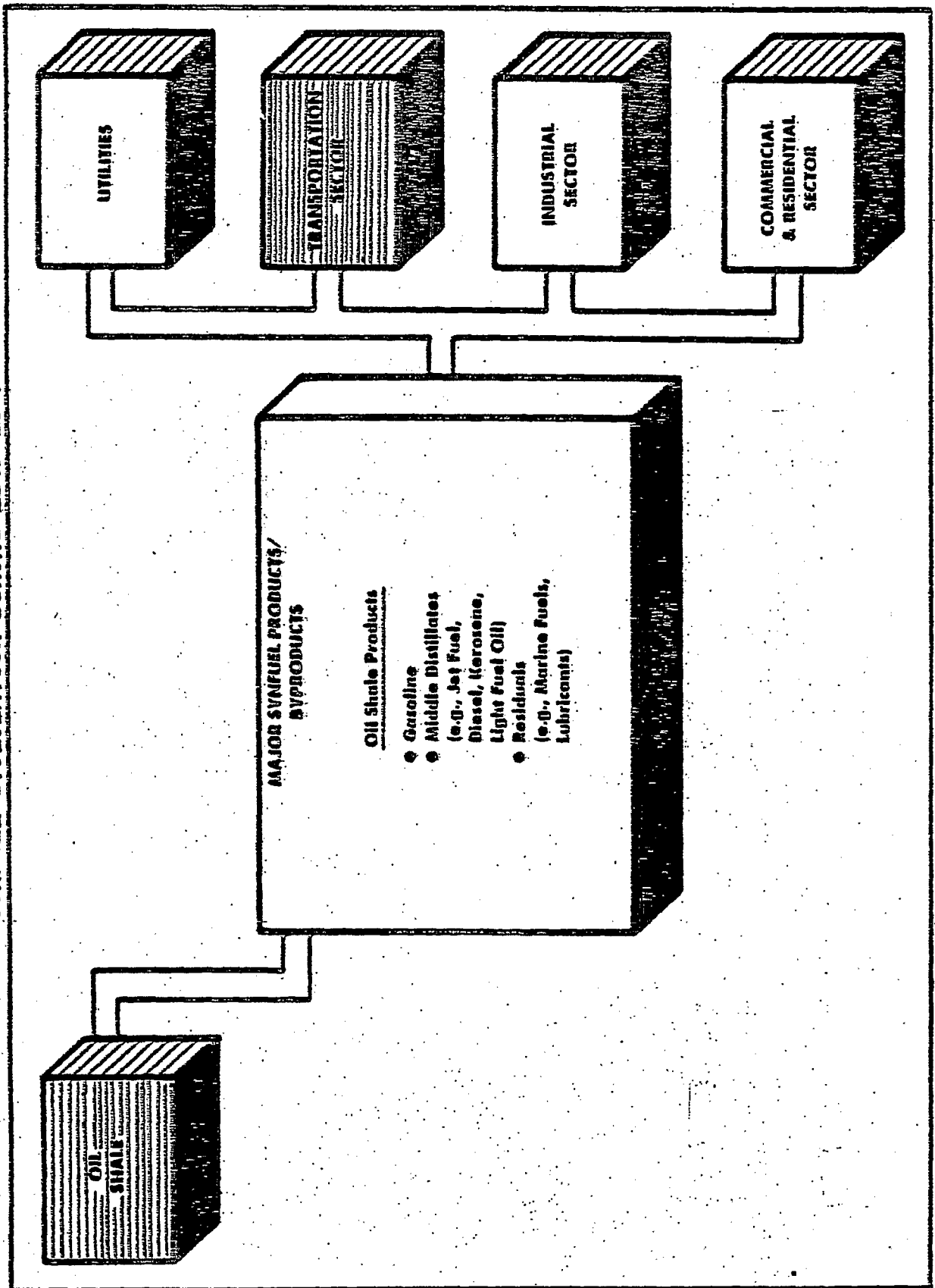
Figure 5

SYNFUEL UTILIZATION DURING 1985-1990

MAJOR SYNFUEL PRODUCTS/ BYPRODUCTS

Oil Shale Products

- Gasoline
- Middle Distillates (e.g., Jet Fuel, Diesel, Kerosene, Light Fuel Oil)
- Residuals (e.g., Marine Fuels, Lubricants)

Coal Gasification Products

- SNG
- Low and Medium Gas
- Methanol
- Petrochemicals

Sectors: UTILITIES, TRANSPORTATION SECTOR, INDUSTRIAL SECTOR, COMMERCIAL & RESIDENTIAL SECTOR

OIL SHALE

COAL GASIFICATIONS

Figure 6

242

SYNFUEL UTILIZATION DURING 1985-2000

MAJOR SYNFUEL PRODUCTS/BYPRODUCTS

Oil Shale Products
- Gasoline
- Middle Distillates (e.g., Jet Fuel, Diesel, Kerosene, Light Fuel Oil)
- Residuals (e.g., Marine Fuels, Lubricants)

Coal Gasification Products
- SNG
- Low and Medium Gas
- Methanol
- Petro-chemicals

Coal Liquefaction Products
- Gasoline
- Middle Distillates (e.g., Jet Fuel, Diesel, Kerosene, Light Fuel Oil)
- Residuals (e.g., Marine Fuels, Lubricants)
- SNG
- Low and Medium Gas
- Methanol
- Petrochemicals

UTILITIES — TRANSPORTATION SECTOR — INDUSTRIAL SECTOR — COMMERCIAL & RESIDENTIAL SECTOR

OIL SHALE — COAL GASIFICATION — COAL LIQUEFACTION

Figure 7

243

power generation by utilities. Some of the major industrial users of high-Btu gas are likely to be textile, food, steel, and chemical industries. Initially, following the current use pattern, it will be used not only as an industrial fuel but also as a chemical feedstock. It is expected that the existing na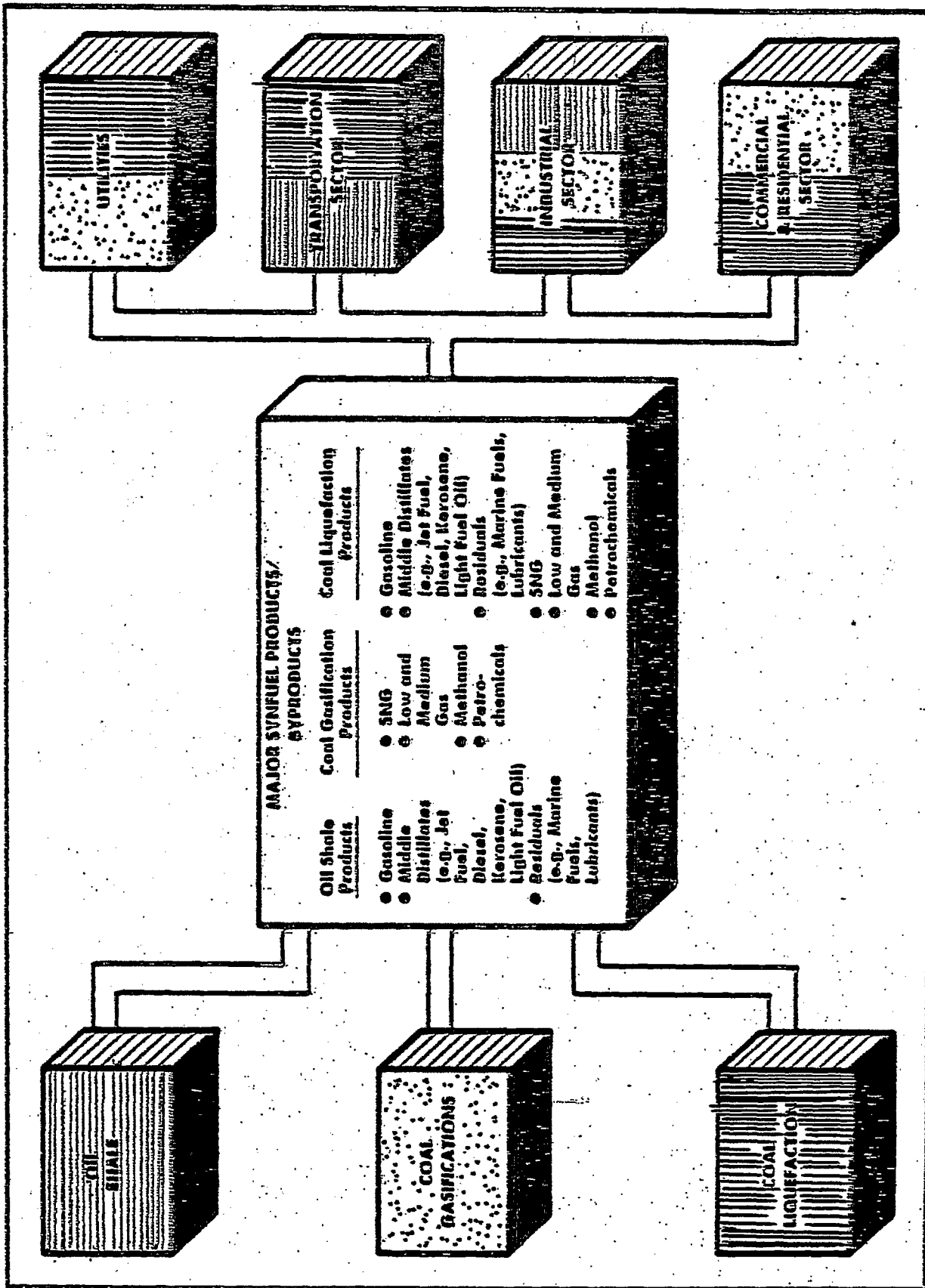tural gas pipeline network, with the exception of a few connecting pipelines, will be utilized for the distribution of high-Btu gas and, therefore, introduction of high-Btu gas is not likely to cause major problems concerning distribution for end-use applications. During this time period it is also likely that low- and medium-Btu gasification plants will be used by industries in a captive mode to supply some of their fuel and chemical feedstock needs. This may amount to as much as 0.3 to 0.4 MMBPD of oil equivalent based on the first two scenarios. The medium-Btu gas could be used as a synthesis gas for the production of different chemical products such as ammonia which in turn could be used for the manufacture of such products as fertilizers, fiber and plastic intermediates, and explosives. Currently the petrochemical industry derives its synthesis gas by reforming natural gas or naphtha. During this time period, it is likely that one to three small plants possibly producing methanol from medium-Btu gas may come on line. These are likely to be owned by industries primarily to supply internal needs. This could be for the production of formaldehyde, a product with a number of end-use applications. It is unlikely that products from these plants will be entering the open market directly, on a large scale, for public consumption. During this time period the use of low-Btu gas will be limited to an industrial fuel in such applications as kilns, chemical furnances and small boilers. However, the use of low-Btu gasification by utilities in one or two demonstration units for combined-cycle applications cannot be ruled out.

During this time frame the shale oil output will continue to grow reaching as much as 0.4 MMBPD in accordance with the National Goal Scenario. As a result it is anticipated that increasing amounts of shale oil products will be entering the transportation sector, with limited entry into the industrial sector for use as fuel.

SYNFUEL PRODUCT UTILIZATION ADDITIONS, 1991-2000

During the 1991-2000 time frame (reference Figure 7), central coal liquefaction plants will introduce into the market a spectrum of products and by-products that will be consumed by the transportation, industrial, and residential/commercial sectors. Based on the nominal and accelerated scenarios, by the year 2000 1.5 to 2.5 MMBPD of coal liquid products will be entering the market. Under these conditions, a significant segment of the transportation fleet could be running on synthetic fuel. Coal-derived liquids will be utilized not only by industry as a fuel source and chemical feedstock, but also by the residential and commercial sectors for space heating, hot water supply and other domestic uses. Furthermore, many of the oil-fired utility plants given exemption from converting to coal in the interim will be burning coal-derived fuel oil. SRC II plants will be the likely candidate which will be supplying the bulk of this fuel. It is also expected that methanol from indirect coal liquefaction could be entering the market for use as turbine fuel for the production of electricity, during this time period. In addition, SNG produced from the liquefaction processes will be also entering the market, supplementing the output from high-Btu gasification plants. The SNG output from liquefaction plants could be as high as 20 percent of the total useful output from these plants in terms of heating value. LPG and naphtha produced from direct and indirect coal liquefaction processes and oil shale are likely to be used primarily by the petrochemical industries. For example, LPG may be used by the petrochemical industry as a raw material for the production of alcohols, organic acids, detergents, plastics, and synthetic rubber components. Naphthas may be utilized for the manufacture of such items as solvents, adhesives, pesticides, and chemical intermediates. Currently the petrochemical industry uses about 11 percent of our crude oil supply for the production of various petrochemicals. During the 1990-2000 time frame, it is possible that the same percentage of available synfuels will be utilized by the petrochemical industry for the production of hundreds of petrochemical products. A major use of residuals from coal liquefaction processes and oil shale is likely to be the manufacture of different types of lubricants. These could be for such applications as lubrication of engines and general machinery, steam turbine bearings and reduction gears, compressors, insulating oils, metal working and cutting oils.

So we see in the above discussion that the synfuels products and by-products are likely to enter all the end-use sectors, in course of time. The potential for exposure and for environmental impacts must be carefully considered. Early planning by the EPA will require that synfuel products/by-products be assessed with regard to their environmental acceptability.

POTENTIAL ENVIRONMENTAL EXPOSURES DUE TO SYNFUELS UTILIZATION

A major concern of the emerging synfuels industry is the potential environmental, health and safety impacts associated with the use of synfuels. The potential exposure of the public to synfuels will depend on the rate of development of the synfuels industry's specific end-use markets. Since the market may cover a wide range of products and end uses, a significant portion of the population may be exposed. The products will enter the markets in varying quantities over the coming years. To illustrate the important environmental concerns, synfuels product production rates based on the National Goals Scenario (Scenario I) are considered and three time periods are examined for potential environmental exposure, 1985-1987, 1988-1990, and 1991-2000.

POTENTIAL EXPOSURES: 1985-1987

During this period, synfuels entering the market will be mostly limited to shale oil products. Approximately 0.2 MMBPD of products by 1987 is projected by Scenario I. Crude shale oil will most likely be transported to refineries in either the Gulf Coast or Midwest and is expected to be distributed by existing pipelines. Product quantities will be limited. The hazards of transporting and storing crude shale oil and shale oil products are expected to be minimal. Shale oil products will be used primarily as transportation fuels such as gasoline, diesel oil, and jet fuel and will be distributed by railroads, tankers, trucks and barges. During this period the quantities handled are estimated to amount to 0.04 MMBPD of gasoline and a combined total of 0.16 MMBPD for the middle distillates. The major exposure to these products occur at storage terminal unloading operations and service station storage tank loading operations, both of which have high spill potential. The end user (a passenger car, truck, or other vehicle) also poses a potential spill problem due to the rapid expansion of self service stations. Combustion of the fuels may expose a large segment of the population since most automobile traffic is generated in central business districts and their suburbs. By-products from shale oil refining such as lubricating oils and greases will be shipped from refinery bulk packing plants in secure containers, minimizing the likelihood of exposure.

Products from shale oil production could reach approximately 0.3 to 0.4 MMBPD during this period, as suggested by the accelerated rate scenario, with the potential exposure reaching twice the level suggested by the National Goal Scenario.

POTENTIAL EXPOSURE: 1988-1990

During this period, in addition to increased shale oil production, SNG low and medium Btu gas, and some indirect liquefaction products will also be entering the market, which increases the complexity of the synfuels distribution network and increases the potential for public exposure to the products. It is a time period by which the EPA must have identified potential problems and have developed a plan for meeting the synfuels challenge.

Shale oil production during this period is projected to be 0.3 to 0.4 MMBPD under the National Goal Scenario, but could range from 0.2 MMBPD (nominal production rate scenario) to 0.8 MMBPD (accelerated production rate scenario) in 1990. The exposure potential to the products will increase proportionally during this time period compared to the previous period.

The SNG entering the market is projected to amount to an oil equivalent of 0.4 MMBPD by 1990, and will be transported by existing pipeline to the various markets. Although pipelines transporting SNG or crude shale oil present a low accident potential, pipelines either transect or terminate in densely populated areas, providing some degree of exposure potential to these products. First-generation coal gasification technology (Lurgi) buildup will occur near western U.S. coal deposits, the Northern Great Plains/Rocky Mountains area. The SNG from this area will enter the northern tier pipeline network and will be distributed across the upper Midwest. Medium- and low-Btu gases will also be in the market during this period, although they will probably be used for internal plant needs. This will minimize the exposure potential since these gaseous products will not require any transportation.

Some synthetic gases have different compositions than natural gas, and may cause internal corrosion and stress-corrosion cracking in pipelines. Effects of impurities on the long-term degradation of some pipeline

materials are unknown. Synthetic gaseous fuels also have different
flammability and explosion limits that may require new techniques in the
management of pipeline leaks. Gases with a high CO concentration are toxic
and could present significant exposure problems.

In addition to their use in transportation and boiler applications,
synfuels products will be used as feedstocks for industrial processes.
These applications, although limited during this time period, present
another avenue of exposure for which EPA must be prepared. The population
exposed could include industrial plant personnel as well as the end users
of the industrial products. During this period, medium-Btu gas could be
used as a synthesis gas for the production of methanol and ammonia, each of
which can be utilized as a finished product.

Although this period will be characterized by the emergence of many
synfuels products, the main population exposure potential will occur from
crude shale oil transport by pipelines, product storage and the combustion
of these products.

A basic environmental concern with the transportation of liquid
synfuels is the possibility of an accidental spill. A recent (1979)
Department of Transportation analysis shows that of all the accidents
resulting from pipelines carrying liquid petroleum products, the largest
spillage occurs from LPG (58.6 percent) followed by crude oil (25.3
percent), with fuel oil (6.1 percent) and gasoline (4.5 percent) being the
other major contributors.

As an example to illustrate the relative exposure of transporting
petroleum products by pipeline in order to provide an awareness of the
potential exposure in transporting shale oil, Table 8 presents a listing of
oil pipeline accidents. Since existing pipelines will be used during this
time period for transporting crude shale oil, these potential exposures and
risks in each component of the carrier system must be considered by EPA.

Table 8. Number of Oil Pipeline Carrier System Accidents

| YEAR	79	78	77	76	75	74	73	72	71
Line Pipe	207	194	177	169	185	203	215	238	264
Pumping Station	20	30	32	11	24	13	23	31	14
Delivery Point	6	4	4	4	5	5	5	3	2
Tank Farm	5	15	12	14	30	22	21	24	11
Other	11	13	12	14	10	13	9	10	19
Total Accidents	249	256	237	212	254	256	273	306	310

Source: Department of Transportation

POTENTIAL EXPOSURE: 1991-2000

This period is characterized by the large-scale entry into the market of direct and indirect liquefaction products and by-products for use primarily by the transportation, industrial and utility sectors. Based on the National Goal Scenario, 1.0 MMBPD of coal liquids will be in the market by 2000, but may range up to 2.5 MMBPD. Utility and industrial boiler fuels produced by coal liquefaction processes will be most in demand in the Gulf Coast, Northeast, and Southern California regions, as shown in Figure 8. These regions contain a significant portion of the U.S. population. The use of these fuels will also have some beneficial effect in areas that are sensitive to particulate and sulfur dioxide since these fuels have lower ash and sulfur contents. As more liquefaction capacity develops in the Appalachian and interior regions, liquid fuels will more readily be used in the industrial areas of Indiana, Illinois, Ohio, and the upper Northwest. Shale oil products during the period may reach a level of 0.4 MMBPD under the National Goal Scenario and could reach as high as 0.9 MMBPD under the accelerated rate scenario. High-Btu gas under these two scenarios is estimated at 0.5 MMBPD and 1.0 MMBPD respectively by 2000. As coal gasification technology develops, it is likely that a key area for gasification will eventually be Appalachia, with SNG entering the existing pipelines and being distributed along the east coast to both industrial and residential users.

This period is also characterized by increased use of coal liquid products for chemical feedstocks and in the housing and commercial sectors

250

Figure 8. Potential Regions for Synfuel Demand for Industrial Fuel Application

High Demand Regions
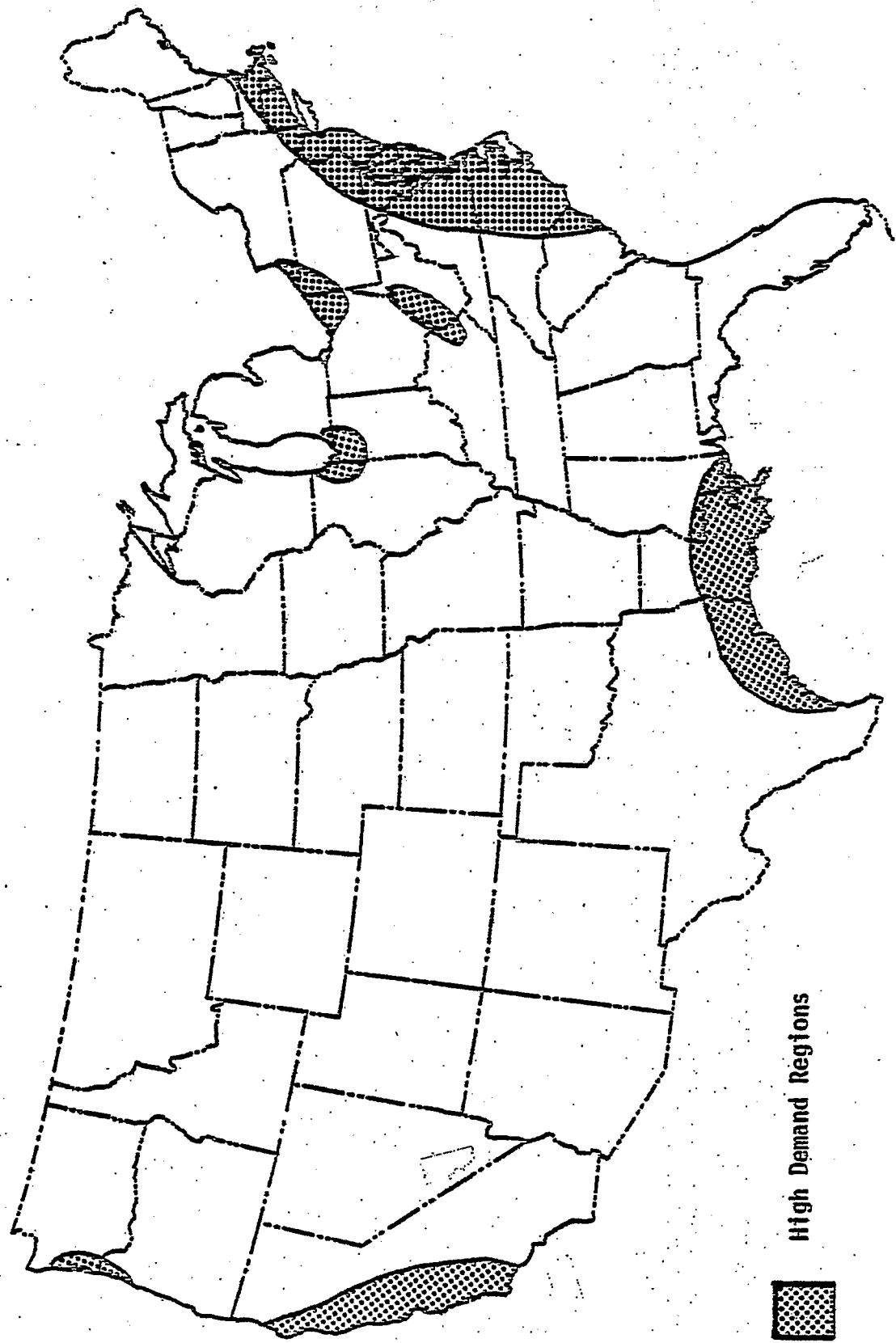
for space heating and hot water supply. Naphthas produced by liquefaction processes are likely to be used by the petrochemical industries for; manufacturing solvents, pesticides and chemical intermediates. Residues from coal liquefaction processes may be used to manufacture several types of lubricants with a wide variety of applications. This market penetration significantly increases exposure potential as there is virtually no segment of the population that would be excluded from the use of synfuel products and by-products.

In addition to synfuels utilization, EPA must also consider the transportation and handling aspects of the synfuels products and by-products. As the synfuels develop during this period, transportation modes other than pipelines will be utilized. Although there are associated risks, pipelines are considered to present less risk than other modes such as railroads, trucks, and tankers. As these modes are currently used for a wide variety of petrochemical products, it is expected that they will also be used as synfuels penetrate the market, thereby presenting another concern that EPA must address.

Table 10 presents an estimate of the range of synfuel products to be shipped by the various transportation modes beginning in the 1990s. Nearer to the year 2000, the relative amounts of products transported between the modes may vary. The majority of the synfuel products as well as crude shale oil will be transported by pipelines, which presents the least amount of exposure potential. On the other hand, railroads which have a high accident potential will transport the least amount of products. In order to supply the high demand regions (reference Figure 8) the transport distribution networks may develop as illustrated in Figure 9. The distribution system indicates that the crude shale oil, refined products, SNG, and coal liquids will each be transported across areas of high population density and industrial concentration, mostly in the eastern U.S. A market for 2.2 MMBPD of synfuels products and by-products by 1992 under the National Goals Scenario indicates the magnitude of the problem for which EPA must prepare.

The transportation modes that will be utilized by the synfuels industry and which pose a greater accident potential than pipeline transport are railroads, trucks and tankers. Railroads will be used
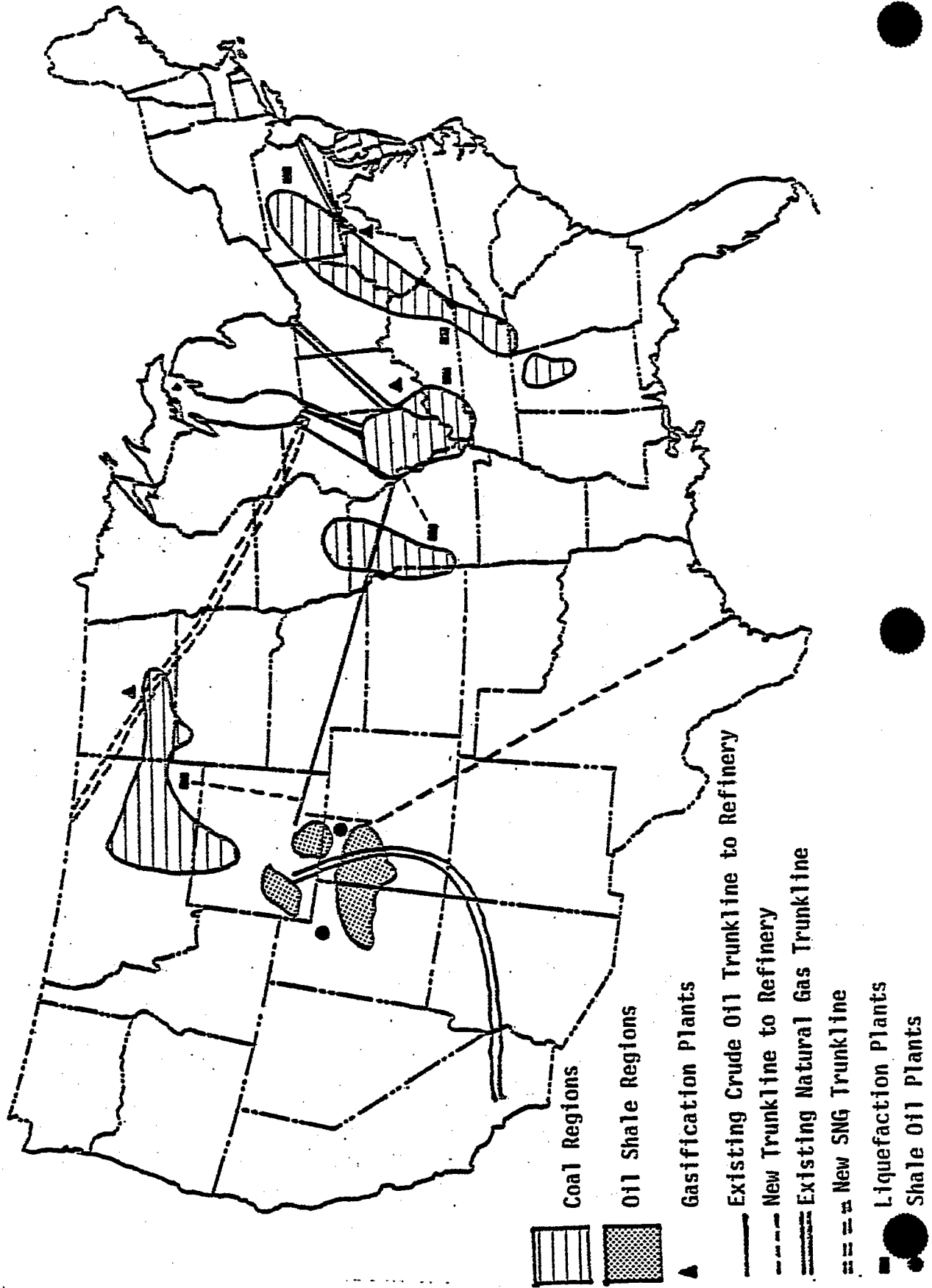
252

Table 10. Range of Synfuels Distributed by Mode of Transportation in the 1990's

SYNFUEL PRODUCT	PIPELINE	RAIL	TRUCK	TANKER OR BARGE
HI Btu GAS (MMSCFD)[2]	1900 – 4800	0	0	0
MEDIUM Btu GAS (MMSCFD)	8300 – 5600	0	0	0
LIQUEFACTION PRODUCTS (MMBPD)				
HEAVY FUEL OILS AND MIDDLE DISTILLATES	0.025 – 0.080	0	0.014 – 0.044	0.007 – 0.022
GASOLINE	0	0	0.072 – 0.520	0.018 – 0.130
NAPHTHA	0	0.042 – 0.067	0.005 – 0.008	0.005 – 0.008
LPG	0	0	0.0 – .018	0.0 – 0.005
CRUDE SHALE OIL (MMBPD)	0.389 – 0.750	0	0	0
REFINED SHALE (MMBPD)				
GASOLINE	0.040 – 0.076	0	0	0.026 – 0.051
JET FUEL	0.047 – 0.090	0	0	0.031 – 0.061
DIESEL OIL	0.084 – 0.162	0	0.084 – 0.162	0.042 – 0.162
RESIDUAL OIL	0.025 – 0.047	0	0.004 – 0.007	0.007 – 0.014
ACCIDENT RISK	LOW	HIGH	HIGH	MODERATE

(1) MMSCFD = Million Standard Cubic Feet per Day

(2) MMBPD = Million Barrels per Day

253

Figure 9. Synfuel Resources and Distribution System in the 1990's

Coal Regions

Oil Shale Regions

▲ Gasification Plants

—— Existing Crude Oil Trunkline to Refinery

— — New Trunkline to Refinery

═══ Existing Natural Gas Trunkline

= = = New SNG Trunkline

■ Liquefaction Plants

● Shale Oil Plants

254

primarily for the transportation of naphthas which in 1992 are estimated to range from 0.04 to 0.07 MMBPD. This mode of transportation presents a high degree of accident risk due to the poor condition of the Nation's rail system. Derailments, grade crossing accidents, and collisions between trains pose potential risks to the transportation of any hazardous or toxic substances. Tank car accidents with hazardous materials are shown in Table 11, providing another example of potential risks associated with transporting synfuels products.

Table 11. Railroad Tank Car Accidents with Hazardous Materials

	1979	1978
Total Accidents	937	1014
Accidents Involving Atmospheric Release	165	228

Source: Federal Railway Administration

The use of tanker trucks will be extensive in transporting coal liquids and refined shale oil products. In 1992 under the National Goal Scenario, approximately 0.4 MMBPD of gasoline from coal liquefaction may be transported by truck, and up to 0.5 MMBPD under the accelerated rate scenario. Other products using this mode are middle distillates and naphthas. Potential exposure to the general population is high with tanker trucks since much of these products will be delivered to urban areas where trucks will face the normal amount of traffic accidents in congested areas. In addition, exposures to the products will occur in loading and unloading of trucks at storage terminals and service stations. Due to vapor recovery requirements mandated by state implementation plans, evaporative emissions of volatile compounds are gradually being controlled, but pollution control systems must be improved to further reduce emissions. Accidents or defective emission control systems provide the chief potential for release of synfuels products by truck transport.

Tankers and barges will also be used for the transportation of the refined shale oil and coal liquids products and could be used extensively if the markets are accessible to the gulf coast. Under the National Goal

255

Scenario, 0.1 MMBPD each of gasoline and diesel oil may be transported by these modes in 1992. Other products to a lesser extent are naphtha, LPG, jet fuel, and residual oil. A significant amount of petrochemical products currently move along the Mississippi River to northern markets. The major emission source for this operation involves loading and unloading; however, the accident rate is less than that of surface transportation mode. As with truck loading, increased emission controls are being initiated for ship and barge loading which will significantly decrease evaporative emissions by the time the synfuels industry is developed. Improvements are also being made to reduce spills of petrochemical products into waterways. Reduction of accidental spills and prevention of intentional releases are currently under regulation by the Coast Guard and EPA.

In addition to transportation and handling, the storage of synfuel products and by-products may pose potential environmental problems. These problems may occur primarily with refined shale oil and coal liquids. As with other petroleum products they will be stored at bulk storage terminals until used. By 1992, a total of 1.4 MMBPD of synthetic liquids will be produced under the National Goal Scenario, and ranging up to 1.7 MMBPD under the accelerated rate scenario. Exposures to these products at the terminals may occur during the loading and unloading operations, as well as breathing losses from the tanks during product storage. The potential for exposure depends upon the volatility of the products and the frequency of loading operations. Since storage facilities are located at refineries, utility and industrial plants, airports and numerous other facilities, exposure potential is significant. Concern over the uncertainties of the constituents of synfuels may lead to storage procedures for these products that are more rigid, and new storage vessels or containers for liquids may be required under stringent specifications. Some emissions may also occur from low-level leakage.

As with other major control requirements for loading and unloading petroleum products, vapor recovery techniques for bulk storage facilities are being improved, primarily by the use of floating roof tanks. Synfuels such as SRC II liquids have vapor pressures similar to No. 6 fuel oil which has very low evaporative emissions and working losses compared to gasoline. Fugitive emissions of synfuels will always be present as they are with

other petroleum products. However, there will be new control systems developed and emissions will be reduced over the next several years through improvements in emissions control procedures in transportation, handling and storage operations. Only after thorough toxicity testing of synfuel products and by-products can an assessment be made of whether synfuels transportation, handling and storage will pose environmental, health, and safety problems greater than those experienced in the petroleum refining and chemical manufacturing industries.

ENVIRONMENTAL CONTROL TECHNOLOGY NEEDED FOR SYNFUELS UTILIZATION

The utilization of synfuels products and by-products will require improvements in existing environmental control technology and the development of new technologies. In order to assess the control technology requirements, it is necessary to first understand the hazards associated with synfuel utilization. This may be accomplished by determining the constituents of synfuels products and by-products, their transformation upon use, and ultimate fate in the environment. These data in turn must be tied closely to the product buildup rate described in each of the scenarios since these impact the types of products produced and their rate of penetration into the market. Once these factors are understood, then control technology options may be evaluated. This cycle must be completed within the next 10 years in order for EPA to meet the synfuels challenge.

EXISTING DATA REGARDING HAZARDS OF SYNFUEL PRODUCTS IS SPARSE

At the current time there is a lack of sufficient data available to properly assess the potential risks associated with the utilization of synfuel products and by-products. The development of these data will require significant efforts on the part of the government, industry and the academic community to generate sound, reliable information to assure minimum risks to the health and welfare of the nation as synfuels are introduced into the market. This synfuels data base must contain not only accurate and representative information about the physical properties, chemical composition and biological activities of synfuels, but must also contain equally comprehensive data on the end uses of the products and by-products.

The DOE and EPA are presently conducting significant research efforts on synfuels product characterization. The results of some of the shale oil and coal liquid products are becoming available. An example of the preliminary analysis of these two products compared with petroleum crude is presented in Table 12. There are some similarities in the diaromatic content between shale oil and petroleum crude, with coal liquids having the highest content. This factor may be significant if a spill of these products occurred, as impacts on water pollution would be less than from coal liquids. A comparison between coal and petroleum derived gasolines is

Table 12. Diaromatic Content of Synthetic Crudes and
Crude Oils

Constituent	Concentration[1], mg/g		
	Typical Shale Oil	Coal Syncrude	Petroleum Crude
Naphthalene	1.39	1.68	0.87
2-Methylnaphthalene	0.91	3.47	1.04
1-Methylnaphthalene	0.68	1.11	0.75
Biphenyl	0.06	0.44	T
2, 6-Dimethylnaphthalene	0.10	0.81	0.08
1, 3/1, 6-Dimethylnaphthalene	1.63	3.01	1.48
2, 3-Dimethylnaphthalene	0.28	1.53	0.51
1, 5-Dimethylnaphthalene	0.03	0.67	0.08
1, 2-Dimethylnaphthalene	0.19	0.23	0.31
Acenaphthalene	0.26	2.19	0.30
Acenaphthene	T	0.30	ND
TOTAL	5.23	15.4	5.42

[1] T = trace, ND = not detected

Source: Oak Ridge National Laboratory

259

presented in Table 13, indicating significant variations in aromatics and unidentified compounds. Due to the high aromatic content of the coal-derived gasoline, potential adverse health effects may occur from widespread use of this fuel in automotive applications. Some of the synfuels products and by-products may be classified as toxic chemicals under the Toxic Substances Control Act (TSCA).

Preliminary health effects studies have indicated that coal liquids have industrial toxicity ratings similar to those of benzoic acid, phosphoric acid, sodium tartrate, and polychlorinated biphenyls (PCB). Coal liquids have also been found to be less toxic than pesticides such as dieldrin and chlordane, and more toxic than crude petroleum and shale oil. Historical epidemiological and animal studies have established that coal tars and pitches from coal coking, gasification, and combustion possess a carcinogenic nature. Although these studies are not all directly comparable, it would appear that some high-boiling point products from direct liquefaction processes or from coal pyrolytic processes may possess a high degree of carcinogenicity.

It is apparent that although work has started in the right direction to assess synfuels hazards, much work still needs to be conducted. As the physical, chemical and biological results are analyzed, and potential risks evaluated, decisions can start to be made as to the various pollution control technologies that can be most effectively applied in the utilization of synfuels.

Table 13. Major Chemical Component Classes of
Petroleum and Coal-Derived Gasoline

CHEMICAL GROUP	GASOLINE	
	Petroleum-Derived[1]	Coal-Derived[2]
Total Saturates	56.38 - 68.68	20.1 - 68.5
Total Alkenes	5.00 - 7.69	0
Total Aromatics	24.32 - 32.91	34.20 - 75.63
Total Unidentified	0 - 3.02	0 - 12.8

[1]Data are from Sanders and Maynard (1968) and Runion (1975).
The range of numbers are for different grades of gasoline
of low, medium, or high octane.

[2]Data are from EPRI (1978). The range of numbers correspond
to different amount of hydroprocessing. Increased hydro-
processing results in fuel with a lower aromatic content.

PRODUCT BUILDUP RATES WILL DETERMINE MAGNITUDE OF ENVIRONMENTAL IMPACTS

Once the hazards of synfuels products and byproducts are known, their relative impacts on the environment will depend upon the product buildup rate and market penetration as described for each of the scenarios. All media, air, water and land, must be considered.

Air pollution impacts will occur primarily from the combustion of synfuels in stationary and mobile sources, with some impacts from fugitive emissions occurring during transportation, storage, and handling operations. Under the National Goal Scenario, coal liquids and shale oil products will contribute the greatest percentage of products. A level of 1.4 MMBPD of these fuels will be produced in 1992 and continue through 2000. Coal liquids will most likely be used in all sectors of the market including utilities, transportation, industrial, and commercial. As most of the products will be used in stationary sources, the air pollution impacts are expected to be less than from shale oil products, all of which will be used by transportation sources. The individual mobile sources do not lend themselves to as effective emission controls as centralized stationary sources. Due to the moderate amount of petroleum product use that is expected to be replaced by synthetic liquids, the air pollution impacts are expected to be moderate.

Under the nominal rate scenario (Scenario 2), only 0.5 MMBPD of liquid fuels will be produced in 1992, and the 1.4 MMBPD level will not be reached until 1998. This will provide relatively lower air pollution impacts from liquids combustion than the National Goal Scenario. The use of low- and medium-Btu gas is projected to be higher under scenario 2 than scenario 1, although air pollution impacts are not considered to be significant since these products will most likely be used for in-plant and feedstock applications.

The greatest relative impact would occur under the accelerated rate scenario, as the quantities of each product are higher than for each of the other two scenarios. By 1992, shale oil and coal liquids production reach a level of 1.8 MMBPD and as much as 3.5 MMBPD by 2000. Shale oil in this period is in excess of 0.9 MMBPD, all of which is used in transportation sources. As shale oil products can be used virtually anywhere in the U.S.,

262

there is very little of the population that may not be exposed to the combustion products. If the majority of the products are used by the military sector, the geographic area of use may be better defined. Significant market penetration under this scenario will also be made by SNG which may be used in all sectors with the exception of transportation. As this product has widespread application, its composition must be accurately defined to determine if combustion will produce air pollution impacts different from use of natural gas.

Water pollution will occur primarily from spills associated with the transportation of synthetic liquids. As the production of these is greatest under the accelerated rate scenario, it provides the greatest potential for these impacts. The crude and refined shale oils, as well as coal liquids will be transported over long distances by pipelines, and then to the markets by various modes of transportation. The loading of tankers and barges, and transportation of the products by waterways provides a moderate degree of spill potential.

Solid wastes will be generated primarily by the pollution control systems used during synfuels utilization. These systems will be limited to stationary source applications where the coal liquids and gases are used in utilities and for industrial processes. As the quantities of solid wastes produced will be dependent on the amount of these fuels used, it will have the greatest impact under the accelerated rate scenario. Oil shale products will not contribute to these impacts since they will be used in transportation sources. By 1992 under this scenario, coal-derived fuels will be produced at a level of 1.8 MMBPD and 4.1 MMBPD by 2000. The majority of these products will be used in stationary sources with emission control systems producing solid wastes. Under scenario 2, only 1.7 MMBPD of coal-derived fuels will be produced by 2000, and 1.8 MMBPD under the National Goal Scenario. As another example of the need to determine synfuel composition, the solid wastes generated by control systems may contain toxic or hazardous components which upon disposal may leach into groundwaters at waste disposal sites.

On the basis of the information presented, significant data need to be developed to assess control technology options. The optimal method of control, if achievable, would be to upgrade the products to remove as much

of the pollutant source content as possible rather than rely on downstream pollution controls. This would have significant benefits on pollution impacts that may occur prior to product utilization. As an example, fugitive emissions into the atmosphere, or spills into waterways would not be expected to be severe if the majority of pollutants were removed during the manufacture of the product.

Once the products are ready for combustion, emission controls will be necessary if product upgrading is unsuccessful. Recent small-scale tests of synfuels combustion have provided encouraging results from an environmental perspective. Several combustion tests of SRC liquids and solids, EDS and H-Coal liquids, shale oil, and coal derived gases have been conducted. For test purposes, some of the combustion devices were not equipped with high-efficiency pollution control devices. Once the products are used in commerce, Best Available Control Technology (BACT) will be required.

EPA is currently proceeding to develop Pollution Control Guidance Documents for all of the synfuel technologies that are being considered under the three scenarios. The purpose of these documents is to foster the development of acceptable synfuels technologies with a minimum of regulatory delays. A similar series of documents may be prepared for the utilization of the products from these technologies.

SIGNIFICANT ENVIRONMENTAL ISSUES FOR SYNFUELS UTILIZATION

- Although a few synfuels products have been included in the toxic substances inventory, most synfuels may be designated as new products under TSCA. EPA will have to identify potential risks associated with the transport and use of synfuels products and by-products, as well as their end uses. Risk and exposure concerns depend on the market infrastructure and likely end use of the variety of products that will result. More diverse end uses and methods of handling, storage, and distribution will increase the exposure potential.

- In addition to TSCA, stipulations of the Clean Air Act will also impact the synfuels market. Atmospheric emissions from fugitive sources are potentially an environmental concern, as well as end-use combustion emissions. These emissions must be characterized so that BACT determinations can be made. Similarities and differences with related petroleum products need to be evaluated.

- Potential atmospheric emissions are much more diverse than the limited set of criteria pollutants which constitute the majority of air pollution concerns today. A critical issue is not so much that hydrocarbons may be an emission, but rather an assessment is needed of the kinds of other organic emissions and the associated risks.

- The potential of accidental spills in the transport and storage of synfuels products and by-products is one of the most critical concerns for protection of groundwater quality and dependent drinking water sources, as stipulated by the Clean Water Act. Additional contamination of receiving waters could be caused by area washdown and stormwater runoff at facilities where minor leakage occurs.

- RCRA requirements will include an integrated solid and hazardous waste management program. Waste oils, storage tank sludges, disposable materials (seals, packing, etc.), and ash residues can all be anticipated from synfuels usage, in addition to waste by-products.

- There is a high probability that synfuels will be blended with petroleum products, either as refinery and petrochemical feeds or as products at end-use locations. EPA will have to judge the applicability of existing regulations covering petroleum product transport and use when the product characterizations are related to blend ratios. Furthermore, synfuels materials that will be used as chemical feedstocks will require environmental assessments regarding their physical, chemical, and biological acceptability.

- The eventual complexity and diversity of the synfuels market infrastructure will represent a challenge to traditional environmental monitoring and inspection procedures, as well as control technology assessment.

● Some of the control approaches will be equipment and operations oriented. This characteristic will require a close EPA interface with other regulatory agencies (such as DOT, ICC, and Coast Guard) regarding transportation operations which are both safe and environmentally acceptable.

● The feasibility of segregating the handling and end-use of potentially hazardous synfuels will certainly have to be evaluated. Proper assessment of environmental risks from synfuels product end-use will be needed to establish exposure estimates.

PERMITTING AND PROGRESS

EPA's regulatory role in an emerging synfuels market will involve permitting for the production, storage, transportation, and end use of the products. Permitting procedures will have to be streamlined to eliminate unnecessary delays in the long-range national goal of reducing petroleum imports. TSCA requirements will be particularly critical in this emerging industry. Plans have been announced by some industries to begin construction of plants to supply SNG and chemical feedstocks. Synfuels projects scheduled for the mid-1980s include shale oil development in Colorado and Utah, and the SRC II demonstration plant in West Virginia. With typical engineering and design efforts requiring 2 years, and construction another 2 to 3 years, it is essential that all permitting be complete within 1 year to keep these critical developments on schedule.