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**Chemical Analysis and
Mutational Assay of Distilled
Oils from the H-Coal Direct
Liquefaction Process:
A Status Report**

B. W. Wilson
D. W. Later
C. W. Wright
D. L. Stewart

January 1985

Prepared for the U.S. Department of Energy
under Contract DE-AC06-76RLO 1830

Pacific Northwest Laboratory
Operated for the U.S. Department of Energy
by Battelle Memorial Institute

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CHEMICAL ANALYSIS AND MUTATIONAL ASSAY OF DISTILLED OILS FROM THE H-COAL DIRECT LIQUEFACTION PROCESS: A STATUS REPORT

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D. W. Later
C. W. Wright
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Pacific Northwest Laboratory
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FOREWORD AND EXECUTIVE SUMMARY

Samples from the H-Coal process, a catalytic, single-stage, coal liquefaction technology, were chemically characterized and screened for microbial mutagenicity. For these investigations, a blend of light and heavy H-Coal process oils was fractionally distilled into 50°F boiling point cuts. The chemical analyses and biological testing results presented in this status report deal primarily with the blended material and the distillate fractions boiling above 650°F.

Results from the microbial mutagenicity assays indicated that onset of biological activity in the crude materials occurred above 700°F. Similar trends have been observed for Solvent Refined Coal (SRC) I, SRC II, Integrated Two-Stage Liquefaction (ITSL) and Exxon EDS process materials. After chemical class fractionation, the primary source of microbial mutagenicity of the crude boiling point cuts was the nitrogen-containing polycyclic aromatic compound (N-PAC) fractions. Amino polycyclic aromatic hydrocarbons (amino-PAH) were present at sufficient concentration levels in the N-PAC fractions to account for the observed mutagenic responses. In general, the chemical composition of the H-Coal materials studied was similar to that of other single-stage liquefaction materials. The degree of alkylation in these materials was determined to be greater than in the SRC and less than in the EDS process distillate cuts.

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Investigations of the H-Coal 50°C boiling point cuts were conducted at Pacific Northwest Laboratory (PNL) by staff members of the Biology and Chemistry Department. These studies were supported by the U.S. Department of Energy, Office of Fossil Energy under Contract DE-AC06-76RLO-1830.

Walter C. Weimer of PNL coordinated the blending and distillation portions of this study. H-Coal materials and sample descriptions were supplied by Wayne Griest of Oak Ridge National Laboratory (ORNL). Distillation of the H-Coal blended oil was performed by Chevron Research Company; R. F. Sullivan authored the report enclosed in Appendix A.

CONTENTS

FORWORD AND EXECUTIVE SUMMARY	iii
CONTRIBUTORS	iv
ACKNOWLEDGMENTS	v
LIST OF FIGURES	viii
LIST OF TABLES	viii
LIST OF ABBREVIATIONS	xi
INTRODUCTION	1
SAMPLE ORIGIN AND METHODOLOGY	3
SAMPLE ORIGIN	3
H-COAL PROCESS DESCRIPTION	3
METHODOLOGY	7
RESULTS	9
CHEMICAL ANALYSES	9
BIOLOGICAL ANALYSES	24
CONCLUSIONS	27
REFERENCES	29
APPENDIX A - CHEVRON DISTILLATION DATA FOR THE H-COAL BLEND AND BOILING POINT CUTS	A.1
APPENDIX B - RAW DATA FOR MUTAGENIC RESPONSE OF <u>S. TYPHIMURIUM</u> WITH H-COAL DISTILLATES AND H-COAL BLENDED MATERIAL	R.1

FIGURES

1. Schematic of the H-Coal Process	6
2. Flow Diagram of the Alumina Adsorption Chromatographic Scheme	8
3. Distribution of Heteroatom Content in H-Coal Distillation Fractions	11
4. High Resolution Gas Chromatogram of the PAH Fraction Isolated from H-Coal Blend. Conditions: 30 m x 0.25 mm i.d.; fused silica capillary column coated with 0.25 μ m film DB-5 (J&W Scientific); 2 min isothermal at 50°C then temperature programmed to 300°C at 3°C/min. Numbered peaks are identified in Table 4	12
5. High Resolution Gas Chromatograms of the PAH Fractions Isolated from H-Coal 50°F Boiling-Point Cuts. Conditions: same as in Figure 4. Numbered peaks are identified in Table 5	14
6. High Resolution Gas Chromatogram of the N-PAC Fraction Isolated from H-Coal Blend. Conditions: same as in Figure 4. Numbered peaks are identified in Table 6	17
7. High Resolution Gas Chromatograms of the N-PAC Fractions Isolated from H-Coal 50°C Boiling Point Cuts. Conditions: same as in Figure 4. Numbered peaks are identified in Table 7	19
8. 12-eV Direct Inlet Probe Mass Spectra of Selected PFP Derivatized Amino-PAH Isolates of the H-Coal 50°F Distillate Cuts: (A) 650-700°F, (B) 700-750°F, (C) 750-800°F and (D) 800-850°F	22

TABLES

1. Distillation and Elemental Analysis Information for the H-Coal Light and Heavy Oil Blended for Distillation	4
2. Distillation Information and Elemental Analysis Data for the H-Coal >650°F Boiling Point Cuts	5
3. Relative Chemical Class Composition of the H-Coal Distillate Fractions	10
4. Quantitative Determinations of Selected Components in the PAH Fraction of the H-Coal Blended Material	13
5. Quantitative Determinations of Selected Components in the PAH Fractions of the H-Coal 50°F Boiling Point Cut Materials	16

6.	Quantitative Determinations of Selected Components in the N-PAC Fraction of the H-Coal Blended Material	18
7.	Quantitative Determinations of Selected Components in the N-PAC Fractions of the H-Coal 50°F Boiling Point Cut Materials	20
8.	Concentrations for Selected Amino-PAH in N-PAC Fractions (A ₃) of the H-Coal Boiling Point Cuts	23
9.	Ratio of Selected Alkylated to Parent Compounds for H-Coal, EDS, and SRC Full-Boiling-Range Materials	24
10.	Ratio of Selected Hydrogenated to Parent Compounds for H-Coal, EDS, and SRC Full-Boiling-Range Materials	24
11.	Mutagenicity of H-Coal Distillates and Blended Material in the Ames Test with <u>Salmonella typhimurium</u> , TA98	26

LIST OF ABBREVIATIONS

AH	aliphatic hydrocarbon(s)
amino-PAH	amino polycyclic aromatic hydrocarbon(s)
bp	boiling point
ECD	electron capture detector
FOB	fuel oil blend
GC	gas chromatography
GC/MS	gas chromatography/mass spectrometry
hydroxy-PAH	hydroxy polycyclic aromatic hydrocarbons
ITSL	integrated two-stage liquefaction
MNNG	N-methyl-N'-nitro-N-nitrosoguanidine
NICI	negative ion chemical ionization
N-PAC	nitrogen-containing polycyclic aromatic compounds
ORNL	Oak Ridge National Laboratory
PAH	polycyclic aromatic hydrocarbon(s)
PDU	process development unit
PFP	pentafluoropropionic
PNL	Pacific Northwest Laboratory
SRC	solvent refined coal

INTRODUCTION

The H-Coal direct liquefaction process is a single-stage technology developed by Hydrocarbon Research Inc. and Cities Service Oil Co. It is based, to a large extent, on H-Oil technology used to convert heavy petroleum resids to lighter products. In contrast to single-stage processes such as solvent refined coal (SRC) I and II, which used no added catalyst in the liquefaction reactor, H-Coal employed a catalyst which was removed and replenished in a batch mode. The process was developed initially between 1964 and 1973, when plans were drafted to use bench-scale and PDU-derived processing data to build a 600-ton/day pilot plant at Catlettsburg, Kentucky. The pilot plant was operated between 1980 and 1982.

Extensive chemical and toxicologic characterization of H-Coal process streams and potential product materials have been carried out by investigators at the Oak Ridge National Laboratory (1-4), and will only be referred to here. The purpose of this document is to report on chemical and toxicologic analyses of materials obtained from a blend of H-Coal middle and heavy distillate, which was fractionally distilled into narrow (50°F) boiling point (bp) cuts. Materials studied here were obtained during operation in the "syncrude" mode.

These investigations were carried out to ascertain the relative chemical and toxicologic properties of H-Coal distillate cuts as a function of distillation temperature, as well as to compare their properties with those of other direct liquefaction materials which have been similarly distilled and analyzed, including blends from the Solvent Refined Coal (SRC) I, SRC II, and Exxon EDS processes. Studies with SRC II materials have shown that mutagenic and carcinogenic activity can be substantially reduced by restricting the upper distillation point of the potential product material to less than 700°F (5). It was of interest to determine whether the H-Coal narrow-cut distillates would exhibit similar properties.

SAMPLE ORIGIN AND METHODOLOGY

SAMPLE ORIGIN

H-Coal materials used in this study were obtained from the Oak Ridge National Laboratory Synfuels Repository. Documentation accompanying the oils indicated that they were from 100-gallon samplings taken on October 30, 1981 from the Catlettsburg, Kentucky plant during the processing of Illinois #6 coal in the syncrude mode. The light oil (N-203 light oil, PNL sample number 3399-146) and the heavy oil (N-202 heavy oil, PNL sample number 3399-147) were blended in a 1.4:1.0 heavy oil:light oil ratio; this blend was representative of a full-boiling-range syncrude material (boiling above 350°F). The blend was then forwarded to Chevron Research Co. in Richmond, California for distillation into narrow-boiling fractions (see Appendix A). Distillation inspection and elemental analysis (carbon, hydrogen, oxygen and nitrogen) data for the oils prior to blending are shown in Table 1. Distillation data for the narrow-range fractions boiling above 650°F are shown in Table 2.

H-COAL PROCESS DESCRIPTION

Figure 1 is a simplified schematic of the H-Coal process (6). Crushed dried coal is slurried with recycle solvent, and hydrogen is added at approximately 200 atmospheres. Slurried material is fed continuously into the ebullated-bed liquefaction reactor. Portions of the reactor catalyst are periodically removed and replaced with fresh catalyst in a batch mode. At the Catlettsburg, Kentucky plant, catalyst replacement was between one and two lb/ton of feed coal. In the syncrude mode, the space velocity in the reactor was nominally 30 lb of coal/hr/cubic foot of reactor volume. Materials produced during syncrude mode operation were of higher hydrogen-to-carbon ratio than would be produced during the fuel oil mode, where the space velocity would be expected to be nominally 80 lb of coal/hr/cubic foot of reactor volume. (The H-Coal pilot plant was operated only in the syncrude mode.) Nominal reactor temperature for both modes was 850°F, and the reactor was operated at a pressure of approximately 3000 psig.

TABLE 1. Distillation and Elemental Analysis Information for the H-Coal Light and Heavy Oil Blended for Distillation

H-Coal Samples		
Coal Type Utilized: Illinois No. 6		
Mode of Operation: Syncrude		
Date of Sampling: 10/30/81		
ASFI Sample Point	210	209
Description	N-203 Light Oil	N-202 Heavy Oil
API Gravity	19.0° - 19.1°	10.1° - 11.1°
C%	86.1	89.0
H%	10.5	9.7
N%	0.57	0.64
S%	0.04	0.07
Volumetric Distillation (°F)		
IBP	344-346	423-458
10	391-392	484-499
30	413-416	532-542
50	437-440	569-579
70	454-457	618-632
90	484-485	710-730
EP	546-627	902-910

Liquid and suspended solid outflow from the reactor are sent to an atmospheric flash separator. The separator overheads are combined with oil from the condenser and sent to an atmospheric pressure still. Overheads from this still constitute the light oil product, and the bottoms are blended with the overheads from the vacuum still; this blend constitutes the heavy distillate product from which the recycle solvent is taken. Bottoms from the atmospheric flash unit are separated in a hydroclone unit to recover oils for the

TABLE 2. Distillation Information and Elemental Analysis Data for the H-Coal
>650°F Boiling Point Cuts

	Boiling Point Cuts, °F				
	650-700	700-750	750-800	800-850	850+
PNL sample identification	5226-094	5226-110	5226-111	5226-112	5226-109
Target boiling range, °F					
Wt% of WOV 4927	6.78	1.36	4.14	1.08	3.09
Lv % WOV 4927	6.47	1.26	3.84	0.98	2.52
Inspections					
Gravity, °API	6.9	1.9	1.4	-2.4	-13.3
Sulfur, wt%	0.32	0.24	0.24	0.24	0.39
Total nitrogen, wt%	0.83	0.76	0.98	1.25	1.80
Oxygen, wt%	1.35	1.47	1.65	0.96	2.73
Hydrogen, wt%	8.92	7.79	7.84	7.48	6.77
Carbon, wt%	88.58	89.74	89.29	90.07	88.31
Distillation, °F	D 1160	ASTM D 1160	ASTM D 1160	ASTM D 1160	TGA ^(a)
St/5	628/655	712/714	738/740	802/804	(705)/818
10/30	656/662	715/719	743/748	807/810	854/919
50	665	722	751	816	961
70/90	666/672	726/734	758/772	824/843	1002/1100
95/EP	674/691	738/755	778/809	856/890	-/1173
% Overhead	99.5	99.0	99.0	99.0	91.0
% in Flask	0.5	1.0	1.0	1.0	9.0
Weighted boiling point, °F ^(b)	3330	3632	3799	4140	
TRP distillation, °F (Corrected from ASTM D 1160 Distillation)					
St/5	599/-	706/-	730/-	793/-	
10/30	653/661	713/718	741/747	804/808	
50	665	722	751	816	
70/90	666/672	726/734	758/772	824/843	
95/EP	-/691	738/755	778/809	856/890	
T*, °F ^(c)	665	712	741	796	856

(a) TGA = Distillation simulated by thermogravimetric analysis.

(b) The weighted boiling point is defined as the sum of 10%, 30%, 70%, 90%, and 95% points from the ASTM distillation (not corrected to TRP).

(c) T* is obtained from the distillation curve (ASTM distillation corrected to true boiling point) in which the boiling point is plotted versus the cumulative liquid volume percent overhead. T* is the intercept of a straight line passing through the 50% point and 30% point with the temperature axis at 0% overhead.

FIGURE 1. Schematic of the H-Coal Process

recycle solvent stream. The remaining solid/liquid stream is sent to a solids/liquid separator. Unreacted carbon and mineral matter leave the process from the liquid/solid separator unit and are not processed further, although designs call for these materials to be utilized eventually as a source of additional hydrogen for the plant.

Vacuum still overheads and bottoms and atmospheric still overheads and bottoms have been toxicologically characterized by Guerin et al. (3). The biological activities of other H-Coal process streams have also been reported (1-4).

METHODOLOGY

Methods used in the characterization and analyses of these H-Coal materials are the same as those used previously to study narrow-range distillate cuts from other processes (5) and will not be discussed in detail here.

Alumina column separations to obtain aliphatic hydrocarbon (AH), polycyclic aromatic hydrocarbon (PAH), nitrogen-containing polycyclic aromatic compounds (N-PAC), and hydroxy-PAH fractions were those described by Later et al. (7,8) and are shown schematically in Figure 2. Gas chromatographic (GC) separations and quantitation as well as GC mass spectral analyses were carried out and evaluated as previously described (5,7-9). High resolution mass spectral analyses were also performed as previously described (10).

Standard plate-incorporation microbial mutagenicity testing using the histidine reversion assay with Salmonella typhimurium, strain TA 98, as previously adapted by Pelroy (10), were performed on the crude distillate cuts as well as on their alumina fractions.

FIGURE 2. Flow Diagram of the Alumina Adsorption Chromatographic Scheme

RESULTS

CHEMICAL ANALYSES

Alumina column separations data on the crude material and the distillate cuts boiling above 650°F are shown in Table 3. N-PAC and hydroxy-PAH content increased with increasing distillation temperature for the H-Coal boiling point (bp) cuts. The concomitant decrease in aliphatic and PAH content noted corresponded to increasing distillation temperature.

Figure 3, taken from inspection data supplied by Chevron Research Co. (see Appendix A), shows the distribution of heteroatoms as a function of boiling point mid-range for the 50°F bp cuts. The relatively high oxygen concentrations below about 500°F were due primarily to the presence of phenols, which distill in this temperature range. Nitrogen content generally increased with molecular weight, as did sulphur concentration. Note that the percentage of molecules in the distillate cuts that has at least one nitrogen atom increases faster than the percent nitrogen itself because the overall molecular weight of the distillate cuts increases with temperature. (Thus, a cut with an average of 18 carbon atoms per molecule would require only about 6% nitrogen by weight to give every molecule, on average, one nitrogen atom. A cut having an average of 6 carbon atoms per molecule would require about three times as much nitrogen to achieve the same average distribution of nitrogen per molecule). This same increase of nitrogen content with molecular weight as a function of distillation temperature has been noted for both SRC and ITSL materials (11).

A gas chromatogram of the PAH fraction isolated from the H-Coal blended material (1.4:1.0 heavy oil:light oil) is shown in Figure 4. The components numbered in this chromatogram are identified and quantified in Table 4. 2-Methylnaphthalene was the component found in the highest concentration in the complex mixture, followed by a component of m/e 168 which was tentatively identified as a dihydrofluorene. Other major components were 1-methylfluorene (which co-eluted with a methyl-substituted dihydrofluorene), naphthalene, phenanthrene, and pyrene. The benzopyrenes were present as minor constituents.

Gas chromatograms of the PAH fractions isolated from the 50°F bp cut materials are shown in Figure 5. There was some overlap of components found in

TABLE 3. Relative Chemical Class Composition of the H-Coal Distillate Fractions

Fraction	Weight Distribution (%) ^(a)					
	H0/L0 Blend (5226-141)	650-700°F (5226-094)	700-750°F (5226-110)	750-800°F (5226-111)	800-850°F (5226-112)	850°F+ (5226-109)
A ₁ : Aliphatic Hydrocarbons	38.9 ± 1.6	15.8 ± 0.5	11.2 ± 0.6	16.4 ± 1.6	16.6 ± 1.2	3.1 ± 0.6
A ₂ : Neutral PAH	36.0 ± 1.6	54.5 ± 0.3	59.1 ± 0.6	50.4 ± 1.9	44.5 ± 0.2	40.1 ± 0.1
A ₃ : N-PAC	8.7 ± 0.2	14.1 ± 0.7	13.9 ± 0.6	15.9 ± 1.2	21.4 ± 4.0	31.4 ± 0.6
A ₄ : Hydroxy-PAH	15.6 ± 0.3	15.3 ± 0.5	15.6 ± 0.2	18.0 ± 0.1	20.0 ± 0.9	25.7 ± 1.2

(a) Values reported are the average of two or three separate analyses and are normalized for 100% recovery.

(b) H0:L0 = Heavy Oil:Light Oil Blend at 1.4:1.0; greater than 350°F full-boiling-range material.

FIGURE 3. Distribution of Heteroatom Content in H-Coal Distillation Fractions

FIGURE 4. High Resolution Gas Chromatogram of the PAH Fraction Isolated from H-Coal Blend. Conditions: 30 m x 0.25 mm i.d.; fused silica capillary column coated with 0.25 μ m film DR-5 (J&W Scientific): 2 min isothermal at 50°C, then temperature programmed to 300°C at 3°C/min. Numbered peaks are identified in Table 4.

TABLE 4. Quantitative Determinations of Selected Components in the PAH Fraction of the H-Coal Blended Material

Peak No. ^(a)	Mol. Wt.	Compound ^(b)	Concentration (parts per thousand)	Peak No. ^(a)	Mol. Wt.	Compound ^(b)	Concentration (parts per thousand)
1	128	Naphthalene	10.3 ± 0.3 ^(c)	22	196, 194	C ₂ -Dihydrofluorenes, C ₂ -Fluorenes	--
2	142	2-Methylnaphthalene	22.8 ± 0.4	23	192	3-Methylphenanthrene	4.56 ± 0.19
3	142	1-Methylnaphthalene	4.33 ± 0.08	24	192	2-Methylphenanthrene	5.14 ± 0.60
4	154	Biphenyl	2.22 ± 0.03	25	192	9- or 4-Methylphenanthrene	1.11 ± 0.47
5	156	2-Ethylnaphthalene	6.71 ± 0.07	26	192	1-Methylphenanthrene	1.98 ± 0.71
6	156	2,6- or 2,7-Dimethylnaphthalene	6.32 ± 0.53	27	206	C ₂ -Phenanthrenes	--
7	156	1,7-Dimethylnaphthalene	2.15 ± 0.02	28	204	Dihydrofluoranthene	2.31 ± 0.03
8	156	1,6- or 1,3-Dimethylnaphthalene	1.55 ± 0.15	29	202	Fluoranthene	1.39 ± 0.40
9	156	2,3- or 1,4-Dimethylnaphthalene	1.96 ± 0.04	30	202	Pyrene	9.03 ± 0.21
10	156	1,2-Dimethylnaphthalene	0.313 ± 0.002	31	204	Dihdropyrene	1.43 ± 0.13
11	154	Acenaphthene	3.16 ± 0.07	32	216	Benzo(a)fluorene	0.653 ± 0.071
12	168	Dibenzofuran	2.00 ± 0.70	33	216	Benzo(b)fluorene ^(g)	4.23 ± 0.39
13	170	C ₃ -Naphthalenes	-- ^(d)	34	218	Dihydro 216	2.20 ± 0.05
14	170	2,3,6-Trimethylnaphthalene	0.790 ± 0.004	35	216	1-Methylpyrene	0.374 ± 0.015
15	166	Fluorene	7.93 ± 0.12	36	230	C ₁ -Pyrenes/fluoranthenes	--
16	168	Dihydrofluorene	20.8 ± 0.28	37	228	Benz(a)anthracene	0.198 ± 0.050
17	168	Dihydrofluorene ^(e)	--	38	228	Chrysene	0.682 ± 0.088
18	180	2-Methylfluorene	5.48 ± 0.96	39	242	6-Methylchrysene	0.668 ± 0.098
19	180	1-Methylfluorene ^(f)	13.0 ± 0.4	40	252	Benzo(j or b)fluoranthene	0.219 ± 0.042
20	182	C ₁ -Dihydrofluorenes	--	41	252	Benzopyrenes	--
21	178	Phenanthrene	9.47 ± 0.86				

(a) Refers to peaks labelled in Figure 4.

(b) Tentative identifications based on retention times and GC/MS data.

(c) Based on three determinations.

(d) No quantitative data.

(e) Co-elutes with m/e 182 (C₁-Acenaphthene) and m/e 184 (C₄-Naphthalene).

(f) Co-elutes with m/e 182 (C₁-Dihydrofluorene).

(g) Co-elutes with 2- or 4-Methylpyrene.

FIGURE 5. High Resolution Gas Chromatograms of the PAH Fraction Isolated from H-Coal 50°F Boiling-Point Cuts. Conditions: same as in Figure 4. Numbered peaks are identified in Table 5.

the materials from adjacent bp cuts. Identification and quantification of the components labeled in Figure 5 are given in Table 5. Dihydrofluoranthene and pyrene were the components found in the highest concentration in the 650-700°F distillate material. Pyrene and benzo(b)fluorene (which co-elutes with 2- or 4-methylpyrene) were found in the highest concentration in the 700-750°F distillate material. Benzo(b)fluorene (or 2- or 4-methylpyrene) and a C₁-substituted benzofluorene and/or a C₂-substituted pyrene, 6- or 4-methylchrysene, and 6- or 4-methylchrysene and benzo(ghi)perylene were the components found in the highest concentration in the 750-800°F, 800-850°F, and 850°F distillate materials, respectively. The benzopyrenes were present in both the 800-850°F and 850°F+ distillate materials.

Figure 6 shows a gas chromatogram of the N-PAC fraction isolated from the H-Coal blended material. Corresponding identification and quantification of the components numbered are given in Table 6. Carbazole, indole, and 3-methylcarbazole were the components found in the highest concentration in this fraction. Alkyl-substituted one-ring aromatic amines were present as major components. Some alkylated phenols were also present as contaminants in the N-PAC fraction due to breakthrough of low-molecular-weight hydroxy-PAH during the alumina column chromatographic step.

Gas chromatograms of the N-PAC fractions isolated from the 50°F bp cut materials from 650°-850°F are shown in Figure 7. Again, as with the PAH fractions, some overlap of components between different distillate materials was evident. The components numbered in Figure 7 are identified and quantified in Table 7. The 3- and 2-methylcarbazoles were the components found in the highest concentration in the 650-700°F distillate material. The components found in the highest concentration in the 700-750°F, 750-800°F and 800-850°F distillate materials were 1- or 4-azapyrene, another C₁-substituted azapyrene (or azafluoranthene), and benzo(c)carbazole, respectively. The 850°F+ distillate material was not analyzed by GC or GC/MS.

Since the N-PAC fractions of the bp cuts were all fairly mutagenic (see biological analysis data), it was of interest to determine whether amino-PAH were present in these isolates and if so, their concentrations. Therefore, the amino-PAH were enriched by silicic acid adsorption chromatography and

TABLE 5. Quantitative Determinations of Selected Components in the PAH Fractions of the H-Coal 50°F Boiling Point Cut Materials

Peak No. (a)	Mol. Wt.	Compound (b)	Concentration (parts per thousand)				
			650 - 700°F	700 - 750°F	750 - 800°F	800 - 850°F	850°F+
1	180	1-Methylfluorene	0.366 ± 0.107 ^(c)				
2	178	Phenanthrene	1.16 ± 0.34				
3	192	3-Methylphenanthrene	6.54 ± 2.22				
4	192	2-Methylphenanthrene	13.3 ± 4.20	0.117			
5	192	9- or 4-Methylphenanthrene	5.01 ± 1.50	--			
6	192	1-Methylphenanthrene	5.00 ± 1.59	--			
7	208	C ₂ -Fluorenes	-- ^(d)	--			
8	204	2-Phenyl naphthalene	22.2 ± 5.20	--			
9	206	C ₂ -Phenanthrenes	--	--			
10	222	C ₂ -Fluorene	--	--			
11	204	Dihydrofluoranthene	33.3 ± 11.1	0.549 ± 0.027			
12	202	Fluoranthene	9.53 ± 2.58	1.81 ± 0.02			
13	218	C ₁ -Phenyl naphthalene	11.0 ± 4.40	--			
14	202	Pyrene	30.0 ± 8.10	99.5 ± 3.4	7.51 ± 0.07		
15	204	Dihydropyrene	4.55	7.68 ± 0.20	0.406 ± 0.006		
16	220	C ₃ -Phenanthrenes	--	--	--		
17	216	C ₁ -Pyrene/fluoranthene	2.25 ± 1.43	14.1 ± 2.1	--		
18	216	Benzo(a)fluorene	--	9.78 ± 2.84	8.04 ± 0.04		
19	216	Benzo(b)fluorene ^(e)	--	83.0 ± 9.4	54.5 ± 3.3		
20	218	Dihydro 216	--	41.5 ± 0.1	20.4 ± 2.0		
21	216	C ₁ -Pyrene/fluoranthene	--	--	--		
22	216	1-Methylpyrene	--	5.78 ± 2.50	5.03 ± 0.52		
23	232	C ₂ -Phenyl naphthalenes	--	--	--		
24	234	Benzonaphthothiophenes	--	--	--		
25	230	C ₁ -Benzofluorenes, C ₂ -Pyrenes ^(f)	--	24.4 ± 1.2	41.3 ± 2.90	0.247 ± 0.008	
26	232	C ₁ -Dihydro 216	--	--	--	--	
27	232	C ₁ -Dihydro 216	--	3.58 ± 0.56	11.2 ± 1.70	0.260 ± 0.016	
28	244	C ₂ -Benzofluorene, C ₃ -Pyrene	--	1.61 ± 0.07	7.90 ± 1.83	1.06 ± 0.02	
29	228	Benzo(a)anthracene	--	0.835 ± 0.121	2.61 ± 1.02	2.35 ± 0.06	0.601 ± 0.043
30	228, 244	Chrysene ^(g)	--	1.88 ± 0.14	11.4 ± 1.4	5.83 ± 0.21	1.33 ± 0.16
31	242	C ₁ -Benz(a)anthracene	--	--	--	--	--
32	258	C ₃ -Benzofluorene, C ₄ -Pyrene	--	--	7.18 ± 0.47	16.5 ± 1.7	1.61 ± 0.11
33	242	3-Methylchrysene	--	--	2.77 ± 0.38	7.81 ± 1.3	1.87 ± 0.27
34	242	2-Methylchrysene	--	--	1.38 ± 0.29	3.14 ± 0.98	0.894 ± 0.155
35	242	6- or 4-Methylchrysene	--	0.546 ± 0.318	6.03 ± 0.21	33.4 ± 0.98	6.27 ± 1.39
36	242	1-Methylchrysene	--	--	--	2.46 ± 0.19	0.759 ± 0.129
37	256	C ₂ -Chrysenes/benzanthracenes ^(f)	--	--	--	1.04 ± 0.54	3.31 ± 0.24
38	272	C ₄ -Benzofluorene, C ₅ -Pyrene	--	--	--	2.47 ± 0.27	0.871 ± 0.127
39	256	C ₂ -Chrysene/benzanthracene	--	--	--	7.43 ± 0.65	2.93 ± 0.18
40	252	Benzo(j or b)fluoranthene	--	--	--	4.34 ± 0.34	4.07 ± 0.59
41	252	Benzo(k)fluoranthene	--	--	--	2.54 ± 1.02	2.32 ± 0.74
42	252	Benzo(e)pyrene	--	--	--	3.31 ± 0.67	3.62 ± 0.21
43	252	Benzo(a)pyrene	--	--	--	2.32 ± 0.12	3.90 ± 0.20
44	276	Indeno(1,2,3-c,d)pyrene	--	--	--	0.384 ± 0.032	3.54 ± 0.68
45	278	Di-benz(a,c or a,h)anthracene	--	--	--	--	1.32 ± 0.28
46	276	Benzo(g,h,i) perylene	--	--	--	0.437 ± 0.033	5.40 ± 0.40

(a) Refers to peaks labelled in Figure 5.

(b) Tentative identifications based on retention time and GC/MS data.

(c) Based on three determinations.

(d) No quantitative data.

(e) Co-elutes with 2- or 4-Methylpyrene.

(f) Quantification is the composite of two peaks.

(g) Co-elutes with C₂-Benzofluorene or C₃-Pyrene.

FIGURE 6. High Resolution Gas Chromatogram of the N-PAC Fraction Isolated from H-Coal Blend. Conditions: same as in Figure 4. Numbered peaks are identified in Table 6.

TABLE 6. Quantitative Determinations of Selected Components in the N-PAC Fraction of the H-Coal Blended Material

Peak No. ^(a)	Mol. Wt.	Compound ^(b)	Concentration (parts per thousand)	Peak No. ^(a)	Mol. Wt.	Compound ^(b)	Concentration (parts per thousand)
1	107	C ₁ -Aniline	2.44 ± 0.33 ^(c)	19	131	2-Methylindole	1.67 ± 0.06
2	107	C ₁ -Aniline	2.72 ± 0.30	20	157	2,6- or 2,7-Dimethylquinoline	4.04 ± 1.18
3	106	C ₁ -Phenol	-- ^(d)	21	157	1,2-Dimethylquinoline	0.578 ± 0.245
4	121	C ₂ -Anilines	--	22	145	2,5- and 2,3-Dimethylindole>	3.39 ± 1.62
5	120	C ₂ -Phenol	--	23	179	Acridine	0.477 ± 0.003
6	133	Tetrahydroquinoline	3.28 ± 0.51	24	179	Benzo(f)quinoline ^(e)	2.65 ± 0.11
7	129	Quinoline	3.97 ± 0.36	25	167	Carbazole	8.66 ± 0.27
8	129	Isoquinoline	0.033	26	193	2- or 3-Methylbenzo(f)quinoline	1.99 ± 0.14
9	135	C ₃ -Aniline	--	27	181	1-Methylcarbazole	2.30 ± 0.09
10	134	C ₄ -Phenol	--	28	181	3-Methylcarbazole	6.26 ± 0.42
11	147	C ₁ -Tetrahydroquinoline	--	29	181	2-Methylcarbazole	4.29 ± 0.45
12	117	Indole	7.26 ± 0.41	30	181	4-Methylcarbazole	1.97 ± 0.05
13	143	2-Methylquinoline	3.74 ± 0.37	31	195	Dimethylcarbazoles	--
14	143	8-Methylquinoline	0.750 ± 0.17	32	203	Azapyrene/fluoranthene	3.76 ± 0.29
15	161	C ₂ -Tetrahydroquinoline	--	33	217	C ₁ -Azapyrene/fluoranthene	--
16	143	7-Methylquinoline	3.49 ± 0.35	34	217	Benzo(a)carbazole	0.814 ± 0.032
17	143	3-Methylquinoline	3.19 ± 0.31	35	217	Benzo(b)carbazole	0.301 ± 0.213
18	131	3-Methylindole	4.38 ± 0.68	36	217	Benzo(c)carbazole	0.808 ± 0.233

(a) Refers to peaks labelled in Figure 6.

(b) Tentative identifications based on retention times and GC/MS data.

(c) Based on three determinations.

(d) No quantitative data.

(e) Co-elutes with phenanthridine.

FIGURE 7. High Resolution Gas Chromatograms of the N-PAC Fractions Isolated from H-Coal 50°C Boiling Point Cuts. Conditions: same as in Figure 4. Numbered peaks are identified in Table 7.

TABLE 7. Quantitative Determinations of Selected Components in the N-PAC Fractions of the H-Coal 50°F Boiling Point Cut Materials

Peak No. (a)	Mol. Wt.	Compound (b)	Concentration (parts per thousand)			
			650 - 700°F	700 - 750°F	750 - 800°F	800 - 850°F
1	179	Acridine	0.385 ± 0.159 ^(c)			
2	179	Benzo(f)quinoline, Phenanthridine	2.07 ± 0.68			
3	167	Carbazole	6.07 ± 1.36			
4	193	2- or 3-Methylbenzo(f)quinoline	14.0 ± 3.8			
5	193	C ₁ -Benzoquinolines	--			
6	181	1-Methylcarbazole	10.7 ± 0.5			
7	193	C ₁ -Benzoquinoline	10.5 ± 4.2			
8	181	3-Methylcarbazole	47.5 ± 1.1	0.294 ± 0.035		
9	181	2-Methylcarbazole	35.1 ± 2.3	0.099		
10	181	4-Methylcarbazole	18.1 ± 1.9	--		
11	211	C ₄ -Azabiphenyls	--	--		
12	207	C ₂ -Benzoquinoline	5.36 ± 2.72	0.312 ± 0.034		
13	207, 195	C ₂ -Benzoquinolines, C ₂ -Carbazoles	--	--		
14	195	C ₂ -Carbazole	6.25 ± 1.82	--		
15	195	C ₂ -Carbazole	11.6 ± 1.7	--		
16	195	C ₂ -Carbazole	9.95 ± 1.42	--		
17	203	1- or 4-Azapyrene		25.7 ± 0.7	22.2 ± 0.3	
18	209	C ₂ -Carbazole		7.75 ± 0.25	--	
19	221, 209	C ₃ -Benzoquinolines, C ₃ -Carbazoles		--	--	
20	223	C ₄ -Carbazoles		--	--	
21	217	C ₁ -Azapyrene/fluoranthene		5.94 ± 0.53	9.67 ± 1.13	
22	217	C ₁ -Azapyrene/fluoranthene		5.53 ± 0.46	6.55 ± 1.90	
23	223	C ₄ -Carbazole		5.56 ± 0.28	--	
24	235	C ₄ -Benzoquinoline		--	--	
25	223	C ₄ -Carbazole		--	--	
26	217	C ₁ -Azapyrene/fluoranthene		4.22 ± 0.24	13.8 ± 1.7	1.17 ± 0.14
27	223	C ₄ -Carbazole		4.50	--	--
28	217	C ₁ -Azapyrene/fluoranthene		--	18.3 ± 2.8	1.65 ± 0.17
29	237	C ₅ -Carbazole		2.81 ± 0.06	6.12 ± 0.41	--
30	231	C ₂ -Azapyrenes (e)			7.07 ± 1.96	0.368 ± 0.003
31	249	C ₅ -Benzoquinoline			--	--
32	231	C ₂ -Azapyrenes			--	--
33	263	C ₆ -Benzoquinolines			--	--
34	217	Benzo(a)carbazole			5.65 ± 1.91	9.17 ± 1.51
35	245	C ₃ -Azapyrene			3.20 ± 0.15	6.51 ± 0.85
36	217	Benzo(b)carbazole			1.37 ± 0.08	9.88 ± 1.81
37	217	Benzo(c)carbazole			2.53 ± 0.46	12.1 ± 0.9
38	231	C ₁ -Benzocarbazole				5.55 ± 0.79
39	231	C ₁ -Benzocarbazoles				

(a) Refers to peaks labelled in Figure 7.

(b) Tentative identifications based on retention time and GC/MS data.

(c) Based on three determinations.

(d) No quantitative data.

(e) Quantification is the composite of three peaks.

derivatized with pentafluoropropionic (PFP) anhydride for selective detection and quantification by probe inlet MS and electron capture detection (ECD) GC (7,12) and negative ion chemical ionization (NICI) GC/MS. Figure 8 shows the probe MS of the enriched, derivatized amino-PAH fractions from the bp cut material. These fractions contained parent as well as alkylated amino-PAH. For example, the 750-800°F distillate cut had major components at m/z 339, 353, 367, 381, 395 and 409 corresponding to the aminophenanthrenes and their C_1 - C_5 alkylated analogues. (Molecular structures of derivatized amino-PAH for other major mass peaks in Figure 8 can be obtained from Table 1 of reference 12.) The major components in most of these derivatized isolates were alkylated amino-PAH. Table 8 provides quantitative data for selected amino-PAH in the five highest-boiling distillate cuts.

Table 9 presents the ratio of amounts of alkylated to parent compounds determined for some selected polycyclic aromatic components which have been quantitated in the PAH and N-PAC fractions of the H-Coal blended material. Data for the same components determined in SRC II fuel oil blend (FOR; 2.9:1.0 middle:heavy distillate), SRC I process solvent, and EDS recycle solvent samples are included for comparison. The H-Coal blended material contained a higher ratio of alkylated parent components than either of the other two single-stage liquefaction products (SRC I and SRC II). The amount of alkylation ranged from 1.2 to 3.9 times greater in the H-Coal material compared to that in the SRC materials, depending on the components chosen. The EDS process material, however, appeared to have a higher ratio of alkylation than the H-Coal material: from 1.2 to 1.6 times greater.

A comparison of the relative amounts of hydrogenated versus parent compounds for the H-Coal blended material, SRC II FOR, SRC I process solvent, and EDS recycle solvent is given in Table 10. For both examples presented, the H-Coal material had a higher ratio of hydrogenation than either of the SRC materials. The EDS recycle solvent had a higher ratio of hydrogenation for dihydrofluoranthene versus fluoranthene, but a lower ratio of hydrogenation for dihydrofluorene versus fluorene.

FIGURE 8. 12-eV Direct Inlet Probe Mass Spectra of Selected PFP Derivatized Amino-PAH Isolates of the H-Coal 50°F Distillate Cuts: (A) 650-700°F, (B) 700-750°F, (C) 750-800°F and (D) 800-850°F

TABLE 8. Concentrations for Selected Amino-PAH in N-PAC Fractions (A₃) of the H-Coal Boiling Point Cuts

Compound ^(a)	Amino-PAH Concentrations in Boiling Pont Cuts (µg/g N-PAC Fraction)				
	650-700°F	700-750°F	750-800°F	800-850°F	850°F+
3-methyl-2-aminonaphthalene	28	---	---	---	---
4-aminobiphenyl	44	---	---	---	---
4-aminofluorene	570	---	---	---	---
2-aminofluorene	764	174	---	---	---
9-aminophenanthrene	---	1,458	42	---	---
3-aminophenanthrene	---	2,915	693	---	---
2-aminoanthracene	---	2,954	1,031	---	---
3-aminofluoranthene	---	291	899	69	---
2-aminopyrene	---	927	---	365	---
1-aminopyrene	---	---	736	413	---
8-aminofluoranthene	---	---	863	3,239	---
6-aminochrysene	---	---	62	311	56
3-aminochrysene	---	---	---	404	177
7-aminobenzo(a)pyrene	---	---	---	62	362

(a) Peak identification assigned by GC ECD retention data and confirmed by NICI GC/MS.

TABLE 9. Ratio of Selected Alkylated to Parent Compounds for H-Coal, EDS, and SRC Full-Boiling-Range Materials

	H-Coal Blended Material	SRC II FOB	SRC I Process Solvent	EDS Recycle Solvent
C ₁ -Phenanthrenes/Phenanthrene	1.35	0.48	0.35	1.61 ^(a)
6- or 4-Methylchrysene/Chrysene	0.98	0.65	0.35	1.59 ^(b)
C ₁ -Carbazoles/Carbazole	1.71	1.44	0.82	1.97 ^(b)

(a) In once-through mode.

(b) In bottoms recycle mode.

TABLE 10. Ratio of Selected Hydrogenated to Parent Compounds for H-Coal, EDS, and SRC Full-Boiling-Range Materials

	H-Coal Blended Material	SRC II FOB	SRC I Process Solvent	EDS Recycle Solvent ^(a)
Dihydrofluoranthene/ Fluoranthene	1.66	0.17	0.22	5.0
Dihydrofluorene/ Fluorene	2.62	--(b)	--(b)	1.33

(a) In bottoms recycle mode.

(b) No m/e 168 (dihydrofluorene) detected.

BIOLOGICAL ANALYSES

The H-Coal blended material, crude 50°F distillates and the chemical class fractions were tested in the standard Ames test with S. typhimurium, TA98. In consideration of previous results from SRC II and EDS coal liquids, which exhibited negligible genetic activity in materials with upper temperature bp

cut points less than 700°F, mutagenicity assays were performed only on the 650-700°, 700-750°, 750-850°, and 850°+F distillate cuts.

Results, summarized in Table 11, are expressed as revertant colonies of S. typhimurium, TA98/μg of test material and also as maximum response in revertants/plate. Mutagenicity potency in revertants/μg was estimated by linear regression analysis of the dose-response data. Raw dose-response data for S. typhimurium, TA98 are presented in Appendix B.

The data in Table 11 illustrate that the mutagenic activity of the unfractionated H-Coal distillates generally tended to increase with increasing distillation temperature. Similar trends in mutagenic activity have been observed with SRC II and EDS coal liquids. However, increases in mutagenic response for the H-Coal distillate cuts appeared to be bimodal, with peak activities in the 700-750°F and 800-850°F distillate fractions. The 650-700°F crude was inactive, confirming that, as with other coal liquids, the biological activity "cut-point" is 700°F. Maximum activity occurred in the 800-850°F distillate.

As seen previously with other 50°F bp cuts of coal liquids, most of the mutagenic activity occurred in the N-PAC fractions and crudes of the distillates above 700°F. Furthermore, as with the unfractionated crude distillate cuts, the mutagenic activity of the N-PAC fractions was bimodal, with peak activities in the same two distillates as for the crudes: 700-750°F and 800-850°F. As was shown in the chemical analysis section, the N-PAC fractions contained significant levels of amino-PAH. Peak concentrations of the 3-ring amino-PAH were observed in the 700-750°F distillate fraction, while peak concentrations of 4-ring amino-PAH were found in the 800-850°F. Most isomers of both the 3- and 4-ring nonalkylated PAH have been shown to be potent mutagens (13). The intermediate bp cut, 750-800°F, contained mostly alkylated 3-ring amino-PAH. Lower concentration levels of highly alkylated 4- and 5-ring amino-PAH were observed in the highest distillate fraction, >850°F.

No genetic activity was observed for the aliphatic fractions. Only the >850°F PAH fraction expressed significant genetic activity. The hydroxy-PAH fractions above 750°F were mutagenically active against S. typhimurium, TA98, with approximately equal activity occurring in the >850°F and 750-800°F distillates.

TABLE 11. Mutagenicity of H-Coal Distillates and Blended Material in the Ames Test with Salmonella typhimurium, TA98

Boiling Point Cut (°F)	Dose Response, 10 ⁷ Revertants/μg ^(a)	Maximum Response in Revertants/Plate ^(b)
<u>Crudes</u>		
850+	70	420±51
800-850	104	481±10
750-800	45	155±19
700-750	65	122±5
650-700	0	56±2

H-Coal blend	11	325±37
<u>Aliphatic Fractions</u>		
850+	0	55±1
800-850	0	60±3
750-800	0	40±1
700-750	0	60±2
650-700	0	69±2

H-Coal blend	0	57±6
<u>Polycyclic Aromatic Hydrocarbon Fractions</u>		
850+	<1	201±3
800-850	0	83±0
750-800	0	81±1
700-750	0	79±0
650-700	0	60±3

H-Coal blend	0	61±2
<u>Nitrogen-Containing Polycyclic Aromatic Compound Fractions</u>		
850+	140	728±22
800-850	340	2096±71
750-800	184	1397±44
700-750	209	1503±8
650-700	109	374±10

H-Coal blend	652	1923±17
<u>Hydroxy-Polycyclic Aromatic Hydrocarbon Fractions</u>		
850+	3 ^(c)	2339±16
800-850	1	377±14
750-800	3	115±13
700-750	0	81±3
650-700	0	65±1

H-Coal blend	2	548±12

- (a) Potency in revertants/μg was derived from the slope of a dose-response curve estimated by linear regression.
- (b) Two sample points (petri plates) were counted to obtain the average maximum response. Each exposure consisted of two points for each concentration assayed. A full dose range consisted of six concentrations.
- (c) Potency estimated on maximum response minus the background, divided by the μg amounts.

CONCLUSIONS

Materials from the H-Coal process were more highly alkylated than those from other single-stage processes such as SRC I and II. The higher alkylation in these materials is probably the result of the catalyst used in the liquefaction reactor. H-Coal materials appeared to be less alkylated than those from the EDS process.

As was the case for the SRC II distillate cuts tested, the mutagenic activity increased with distillation temperature for cuts boiling above 650°F, with maximum mutagenic activity occurring in the 800-850°F cut. Work is now in progress to determine the tumor initiating activity of these higher-boiling distillation cuts in the initiation/promotion (I/P) mouse skin assay. The slight drop-off in mutagenic activity for the highest-boiling H-Coal distillate cut was also observed for the SRC II materials (5). However, when the SRC materials were tested for initiation of mouse skin tumorigenicity and mouse skin carcinogenicity in the chronic skin painting assay, the roll-off at higher distillation temperatures was not observed (10). As with 50°F bp cuts of other process distillates, the mutagenic activity of these materials in S. typhimurium, TA98, was primarily due to the N-PAC components, particularly the amino-PAH.

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APPENDIX A

CHEVRON DISTILLATION DATA FOR THE H-COAL BLEND AND BOILING POINT CUTS

REFINING AND UPGRADING OF SYNFUELS
FROM COAL AND OIL SHALES
BY ADVANCED CATALYTIC PROCESSES

Eighth Interim Report
Task 11: Distillation of H-Coal
Into Narrow Boiling Fractions

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Richmond, California

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I. Abstract

About 55 gal. of a 400°F+ blend of products from the H-Coal Liquefaction Process was distilled into narrow boiling fractions of 50°F increments. These samples were sent to Battelle Pacific Northwest Laboratories for health-related studies.

II. Contract Objective and Status

The objective of this program is to determine the feasibility and to estimate the costs of hydroprocessing synthetic crude feedstocks to distillate fuels using modern commercial petroleum processing technology. Tests are conducted only to the extent needed to enable making reasonable estimates of commercial plant performance and only to the extent a commercial plant is feasible using presently existing technology, subject to the mutual agreement of DOE and Chevron Research. Tests are made for each whole synthetic oil and, where appropriate, for the fractions derived therefrom. Tests will not be carried out for processes which can be reliably estimated.

The feedstocks under this contract and the status of work on each are as follows:

1. Work on the Paraho shale is complete and reported in the first interim report, FE-2315-25, April 1978. Refining cost for shale oil is updated in the fourth interim report, FE-2315-50, June 1980.
2. Work on SRC-I is complete and reported in the second interim report, FE-2315-45, November 1979.
3. Work on SRC-II process product derived from Pittsburgh Seam coal is complete and reported in the third interim report, FE-2315-47, March 1980. (This report was reissued with revisions in November 1982.)
4. Work on H-Coal process product derived from Illinois No. 6 is complete and reported in the fifth interim report, DOE/ET/10532-T3 (FE-2315-61) (DE 82001127), September 1981. An addendum to this report, FE-2315-77, was issued in May 1982.
5. Work on H-Coal process product derived from Wyodak coal is complete. Results are also reported in DOE/ET/10532-T3 (FE-2315-61) (DE 82001127), September 1981.
6. The pilot plant program with EDS process product from Big Brown, Texas, lignite is complete; engineering studies are being completed, and an interim report is being prepared.
7. At the request of DOE, we added Task 9 to the contract. In this task we prepared hydrotreated SRC-II at several severities to supply DOE samples for biological studies. The pilot plant program is complete and the product has been sent to ORNL. Results are reported in DOE/ET/10532-T5 (FE-2315-74) (DE 82013235), April 1982.

8. At the request of DOE, we added Task 10 to the contract. In this task we hydrotreated EDS and H-Coal to supply DOE with product for biological studies. Results are reported in FE-2315-84, issued in January 1983 in final form.

9. At the request of DOE, we added Task 11 to the contract. In this task we distilled a barrel of H-Coal process product into 50°F boiling fractions to supply Battelle Northwest Laboratories with samples for health-related studies. This report completes work under this Task.

10. At the request of DOE, we added Task 12 to the contract. In this task we are upgrading process product from the ITSL process for distillate fuels. The pilot plant tests are in progress.

III. Task 11: Distillation of H-Coal Product Blend for Battelle Pacific Northwest Laboratories

In Task 11, we distilled a blend of products from the H-Coal Liquefaction Process into narrow boiling fractions of 50°F increments. These samples were sent to Battelle Pacific Northwest Laboratories for health-related studies.

One 55-gal. drum blend of H-Coal middle and heavy distillate (blending ratio was 40 wt % middle distillate to 60 wt % heavy distillate) was received by Chevron Research from Battelle on November 8, 1982. The sample was analyzed by Chevron and analyses are reported in Table I. Figure 1 shows the distillation curve for this blend.

DOE asked that we hold a 1-gal. retain of the blend for their future use. Except for this required retain, the blend was distilled in a large batch still filled with Penn State packing. The blend and the distilled fractions were always handled under a nitrogen atmosphere. The reflux ratio for the distillation was 5/1. A "start"-400°F fraction and 400-450°F fraction were removed at atmospheric pressure. The remainder of the distillation was carried out at 10 mm pressure. Fractions were distilled in 50°F increments to 700°F. One gal. of each of the overhead samples was shipped to Battelle on March 1. Distillation yields are given in Table II. Figure 2 shows that a sharp separation was obtained for the seven fractions boiling below 700°F. Inspections of the fractions are given in Table III.

The 700°F+ bottoms (about 5 gal.) from the distillation described in the preceding paragraph was further distilled in a 2-in. by 18-in. crude column with three trays. The latter distillation was performed at 1 mm pressure. Four fractions were obtained: a small amount of 700°F- material, two higher boiling cuts, and the bottoms boiling above about 850°F. The material balance for the second distillation Table IV. Analyses of the fractions from the second distillation are given in Table V.

The distillations of the individual fractions show that the second distillation was not very sharp. Therefore, Fractions 2 and 3 (identified as SGQ 8392 and SGQ 8393) were redistilled in a 2-in. by 18-in. column filled with Penn State packing for better fractionation. The column was operated at 1 mm pressure. The target boiling ranges for the three fractions obtained were 700-750°F, 750-800°F, and 800-850°F. The 850°F+ bottoms could not be distilled further without signs of decomposition (cracking); so no higher boiling distillate fractions were obtained. Table I shows the material balance, the ASTM distillations for the overhead fractions, a simulated distillation of the bottoms,

and other analytical information. Figure 1 shows the distillation curves of the fractions. Figure 2 is a plot showing the heteroatom content of the individual fractions prepared. The results show that the fractionation was sharp, considerably better than obtained when the 700°F+ material was distilled previously. The fractions appear suitable for Battelle's purpose.

Except for quantities necessary for a few key inspections, the fractions were shipped to Battelle for their health-related studies.

Work under Task 11 is now complete.

:fes

REFINING AND UPGRADING OF SYNFUELS
FROM COAL AND OIL SHALES
BY ADVANCED CATALYTIC PROCESSES
CONTRACT NO. DE-AC22-76ET10532

Index of Enclosures

<u>Tables</u>	<u>Title</u>	<u>Drawing No.</u>
I	Analyses of Blend of H-Coal Products Received from Battelle Pacific Northwest Laboratories	RE 832135
II	Distillation of H-Coal Blend, WOW 4927, Distillation No. 103-99	RE 832136
III	Distillation of H-Coal Oil Blend for Battelle Northwest Laboratories (Distillation No. 103-99)	RD 832137-1
IV	Distillation of 700°F+ Fraction from H-Coal Blend, WOW 4927, Distillation No. ASL 1273	RE 832138
V	Distillation of 700°F+ Bottoms from H-Coal Oil Blend into Narrow Boiling Fractions for Battelle Pacific Northwest Laboratories (Distillation No. ASL 1273)	RE 833958
VI.	Distillation of 700°F+ H-Coal Oil into Narrow Boiling Fractions (Distillation Numbers ASL 1273, 1446, and 1447)	RE 833959

Figures

1	Simulated TBP Distillation of H-Coal Oil Blend (WOW 4927) Received from Battelle	RE 831699
2	ASTM Distillations (Corrected to TBP) of Narrow Boiling H-Coal Oil Fractions from Distillation 103-99	RE 831700-1
3	ASTM D 1160 Distillations (Corrected to TBP) of Narrow Boiling H-Coal Oil Fractions	RE 833981
4	Distribution of Heteroatom Content of H-Coal Oil Fractions	RE 833982

TABLE I

DOE CONTRACT DE-AC22-ET10532
 ANALYSES OF BLEND OF H-COAL PRODUCTS
RECEIVED FROM BATTELLE PACIFIC NORTHWEST LABORATORIES

Chevron Identification No.	WOW 4927
Inspections	
Gravity, °API	12.6
Sulfur, Wt %	0.82
Total Nitrogen, Wt %	0.60
Basic Nitrogen, Wt %	0.45
Oxygen, Wt %	1.21
Hydrogen, Wt %	9.33
Carbon, Wt %	88.04
Chlorine, ppm	43.0
Iron, ppm*	76.2
Ramsbottom Carbon, Wt %	0.69
Ash, %	0.01
Hot Heptane Insolubles, %	0.20
Viscosity, cSt at 100°F	5.60
Bromine No.	52.0
Maleic Diene No., cg I ₂ /g	7.44
Water, %	0.26
TBP Distillation, °F	
(Simulated by Chromatography)	
St/5	366/401
10/30	414/473
50	527
70/90	588/694
95/99	751/899
Molecular Weight (Estimated by Chromatography)	229
Molecular Weight (Analyzed)	203
Pour Point, °F	-15

*By atomic adsorption. Other metals detected:

Calcium, ppm	0.2
Chromium, ppm	0.2
Nickel, ppm	1.0
Silicon, ppm	0.3
Zinc, ppm	4.6

TABLE II

DOE CONTRACT DE-AC22-76ET10532
 DISTILLATION OF H-COAL BLEND, WOW 4927,
 DISTILLATION NO. 103-99*

Chevron I.D.	Fraction No.	Boiling Range, °F	Product Wt, Lb	Wt %	LV %	Gravity, °API
SGQ 8384	1	Start-400	19.8	5.84	6.16	21.3
SGQ 8385	2	400-450	55.3	16.31	16.86	18.3
SGQ 8386	3	450-500	53.0	15.63	16.08	17.6
SGQ 8387	4	500-550	56.0	16.51	16.90	16.8
SGQ 8388	5	550-600	59.0	17.40	17.25	12.2
SGQ 8389	6	600-650	40.0	11.80	11.63	11.3
SGQ 8390	7	650-700	23.0	6.78	6.47	6.9
-	8	700+**	<u>33.0</u>	<u>9.73</u>	<u>8.65</u>	-2.7
		Total	339.1	100.00	100.00	

*Distillation in large batch still, at 5:1 reflux ratio, using column with Penn State packing. First two fractions removed at atmospheric pressure, remainder of distillation at 10 mm.

**The 700°F+ fraction was further distilled in another column to obtain the higher boiling fractions. (See Table II.)

TABLE III

DOE CONTRACT DE-AC22-76ET10532
DISTILLATION OF H-COAL OIL BLEND FOR BATTELLE NORTHWEST LABORATORIES
(DISTILLATION NO. 103-99)

Sample Identification	SGQ 8384	SGQ 8385	SGQ 8386	SGQ 8387	SGQ 8388	SGQ 8389	SGQ 8390
Boiling Range, °F	St-400	400-450	450-500	500-550	550-600	600-650	650-700
Wt % of Blend	5.84	16.31	15.63	16.51	17.40	11.80	6.78
LV % of Blend	6.16	16.86	16.08	16.90	17.25	11.63	6.47
Inspections							
Gravity, °API	21.3	18.3	17.6	16.8	12.2	11.3	6.9
Sulfur, Wt %	0.13	0.11	0.12	0.090	0.094	0.24	0.32
Total Nitrogen, Wt %	0.65	0.56	0.44	0.52	0.39	0.60	0.83
Oxygen, Wt %	2.79	4.01	1.73	0.48	0.39	0.51	1.35
Hydrogen, Wt %	10.36	9.88	9.91	10.22	9.45	9.52	8.92
Carbon, Wt %	86.07	85.44	87.80	88.69	89.68	89.14	88.58
Group Type, LV %	Low Mass	High Mass	High Mass	High Mass	22-Component	22-Component	22-Component
Paraffins	11.5	4.3	4.1	4.8	6.3	8.6	4.9
Naphthenes	28.5	19.7	24.9	23.4	8.7	8.1	4.9
Aromatics	60.1	76.0	71.0	71.8	83.2	77.6	78.5
Sulfur Compounds	-	-	-	-	1.7	5.7	11.6
ASTM Distillation, °F	D 86	D 86	D 86	D 86	D 86	D 86	D 1160
St/5	254/374	410/414	452/455	492/492	544/550	593/599	628/655
10/30	376/380	415/417	456/458	496/498	551/554	602/605	656/662
50	382	419	460	502	557	608	665
70/90	384/390	422/428	462/468	506/511	560/566	610/615	666/672
95/EP	394/438	430/450	471/484	512/526	570/575	618/623	674/691
% Overhead	99	99	99	98	99	98.5	99.5
% in Flask	1	1	1	2	1	1.5	0.5
Weighted Boiling Point, °F ¹	1924	2112	2315	2523	2801	3050	3330
TBP Distillation, °F (Corrected from ASTM Distillation)							
St/5	251/-	409/-	456/-	498/-	552/-	606/-	599/-
10/30	379/388	422/427	466/471	508/513	569/576	627/634	653/661
50	392	431	475	521	582	640	665
70/90	396/405	437/446	479/488	528/536	588/597	644/652	666/672
95/EP	-/457	-/472	-/507	-/554	-/608	-/662	-/691
T*, °F ²	382	421	465	501	567	625	665

¹The weighted boiling point is defined as the sum of 10%, 30%, 70%, 90%, and 95% points from the ASTM distillation (not corrected to TBP).

²T* is obtained from the distillation curve (ASTM distillation corrected to true boiling point) in which the boiling point is plotted versus the cumulative liquid volume percent overhead. T* is the intercept of a straight line passing through the 50% point and 30% point with the temperature axis at 0% overhead.

TABLE IV

DOE CONTRACT DE-AC22-76ET10532
DISTILLATION OF 700°F+ FRACTION
FROM H-COAL BLEND, WOW 4927,
DISTILLATION NO. ASL 1273*

Chevron I.D.	Fraction No.	Nominal Boiling Range, °F	Product Wt, g	Wt % of WOW 4927	LV % of WOW 4927	Gravity, °API
SGQ 8391	1	Start-700	80	0.06	0.05	3.4
SGQ 8392	2	700-750	4297	3.06	2.84	2.4
SGQ 8393	3	750-800	4914	3.52	3.24	1.9
SGQ 8394	4	800+	3819	3.09	2.52	-13.3

*Distillation in 18-in. x 2-in. crude column with three trays. Entire distillation at 1 mm pressure.

TABLE VI

DOE CONTRACT DE-AC22-76ET10532
DISTILLATION OF 700°F+ H-COAL OIL
INTO NARROW BOILING FRACTIONS
(DISTILLATION NUMBERS ASL 1273, 1446, AND 1447)

Sample Identification	SGQ 8848	SGQ 8849	SGQ 8850	SGQ 8394
Target Boiling Range, °F	700-750	750-800	800-850	850+
Wt % of WOW 4927	1.36	4.14	1.08	3.09
LV % of WOW 4927	1.26	3.84	0.98	2.52
Inspections				
Gravity, °API	1.9	1.4	-2.4	-13.3
Sulfur, Wt %	0.24	0.24	0.24	0.39
Total Nitrogen, Wt %	0.76	0.98	1.25	1.80
Oxygen, Wt %	1.47	1.65	0.96	2.73
Hydrogen, Wt %	7.79	7.84	7.48	6.77
Carbon, Wt %	89.74	89.29	90.07	88.31
Distillation, °F	ASTM D 1160	ASTM D 1160	ASTM D 1160	TGA ¹
St/5	712/714	738/740	802/804	(705)/818
10/30	715/719	743/748	807/810	854/919
50	722	751	816	961
70/90	726/734	758/772	824/843	1002/1100
95/EP	738/755	778/809	856/890	-/1173
% Overhead	99.0	99.0	99.0	91.0
% in Flask	1.0	1.0	1.0	9.0
Weighted Boiling Point, °F ²	3632	3799	4140	
TBP Distillation, °F (Corrected from ASTM D 1160 Distillation)				
St/5	706/-	730/-	793/-	
10/30	713/718	741/747	804/808	
50	722	751	816	
70/90	726/734	758/772	824/843	
95/EP	738/755	778/809	856/890	
T*, °F ³	712	741	796	856

¹TGA = Distillation simulated by thermogravimetric analysis.

²The weighted boiling point is defined as the sum of 10%, 30%, 70%, 90%, and 95% points from the ASTM distillation (not corrected to TBP).

³T* is obtained from the distillation curve (ASTM distillation corrected to true boiling point) in which the boiling point is plotted versus the cumulative liquid volume percent overhead. T* is the intercept of a straight line passing through the 50% point and 30% point with the temperature axis at 0% overhead.

TABLE V

DOE CONTRACT DE-AC22-76ET10532
DISTILLATION OF 700°F+ BOTTOMS FROM H-COAL OIL BLEND
INTO NARROW BOILING FRACTIONS FOR
BATTELLE PACIFIC NORTHWEST LABORATORIES
(DISTILLATION NO. ASL 1273)*

Sample Identification	SGQ 8391	SGQ 8392	SGQ 8393	SGQ 8394
Nominal Boiling Range, °F	St-700	700-775	775-850	850+
Wt % of Blend	0.06	3.06	3.52	3.09
LV % of Blend	0.05	2.84	3.24	2.52
Wt of Sample, g	84	4542	5212	4572
Inspections				
Gravity, °API	3.4	2.4	1.9	-13.3
Sulfur, Wt %	0.23	0.24	0.24	0.39
Total Nitrogen, Wt %	0.82	0.84	1.12	1.80
Oxygen, Wt %	0.88	1.47	1.50	2.73
Hydrogen, Wt %	8.45	8.13	8.00	6.77
Carbon, Wt %	89.62	89.32	89.14	88.31
Group Type, LV %				
Paraffins	3.5	3.4	2.7	NA
Naphthenes	4.4	3.2	3.1	NA
Aromatics	82.5	82.2	84.4	NA
Sulfur Compounds	9.6	11.1	9.7	NA
ASTM Distillation, °F		ASTM D 1160	ASTM D 1160	TGA**
St/5		713/723	736/750	705/818
10/30	NA	726/730	754/761	854/919
50		734	770	961
70/90		740/755	784/810	1002/1100
95/EP		767/802	832/861	-/1173
% Overhead		99	99	91
% in Flask		1	1	9
TBP Distillation, °F (Corrected from ASTM Distillation)				
St/5		697/-	712/-	
10/30		724/729	749/758	
50		734	770	
70/90		744/755	784/810	
95/99		767/802	832/861	
Ramsbottom Carbon, Wt %	NA	0.38	0.52	19.19
Hot Heptane Asphaltenes, Wt %	NA	0.14	1.51	NA

NA = not analyzed

TGA = thermogravimetric analysis

*This distillation did not appear sharp enough for Battelle's purposes.
Therefore, further distillations were performed on SGQ 8392 and SGQ 8393.

FIGURE 1

DOE CONTRACT DE-AC22-76ET10532
SIMULATED TBP DISTILLATION OF
H-COAL OIL BLEND (WOW 4927)
RECEIVED FROM BATTELLE

FIGURE 2

DOE CONTRACT DE-AC22-76ET10532
ASTM DISTILLATIONS* (CORRECTED
TO TBP) OF NARROW BOILING H-COAL
OIL FRACTIONS FROM DISTILLATION 103-99

*Distillation of SGQ 8390 is by ASTM D 1160,
Others are by ASTM D 86

FIGURE 3

DOE CONTRACT DE-AC22-76ET10532
ASTM D1160 DISTILLATIONS (CORRECTED
TO TBP)* OF NARROW BOILING H-COAL OIL FRACTIONS

*Distillation of SGQ 8394 is by a thermogravimetric method.

FIGURE 4

DOE CONTRACT DE-AC22-76ET10532
DISTRIBUTION OF HETEROATOM CONTENT OF
H-COAL OIL FRACTIONS

TABLE B1. Averaged Positive and Negative Control Data for Salmonella typhimurium TA98 for the Dose-Response Data of Table B2

Compound	µg/Plate	S9 (ml)	Number of Average Revertant Colonies/Plate
2-Aminoanthracene	1.0	10	2675 ± 642
Benzo[a]pyrene	10	100	301 ± 54
MNNG ^(a)	1.0	0	27 ± 11
030 A ₃ ^(b)	1.0	20	167 ± 83
None	0	20	45 ± 10
None	0	20	468 ± 42 (LL) ^(c)
None	0	100	45 ± 14
None	0	100	458 ± 41 (LL)
None	0	0	36 ± 8
None	0	0	453 ± 66 (LL)

(a) MNNG = N-methyl-N'-nitro-N-nitrosoguanidine

(b) 030 A₃ = SRC II nitrogen-containing polycyclic aromatic compounds, 850+°F

(c) LL = Large lens

TABLE B2. Reverse Mutation Dose-Response Data for Salmonella typhimurium in a Standard Ames Plate Incorporation Assay

Boiling Point Cut	Fraction	S9 (μ l)	μ g/Plate	Revertant Colonies TA98/ μ g	
H-Coal Blended (5226-141)	A ₁	100	0	55 $\pm$	5
			20	55 $\pm$	1
			40	42 $\pm$	3
			80	43 $\pm$	3
			100	45 $\pm$	2
			200	39 $\pm$	4
			500	51 $\pm$	6
	A ₂	100	0	55 $\pm$	5
			20	34 $\pm$	1
			40	43 $\pm$	3
			80	51 $\pm$	0
			100	61 $\pm$	2
			200	52 $\pm$	4
			500	51 $\pm$	1
	A ₃	20	0	473 $\pm$	4
			0.2	471 $\pm$	1
			0.4	629 $\pm$	14
			0.8	849 $\pm$	6
			1.0	1036 $\pm$	20 ^(a)
			2.0	1652 $\pm$	178
			5.0	3836 $\pm$	41
	A ₄	100	0	55 $\pm$	5
			20	83 $\pm$	3
			40	155 $\pm$	1
			80	260 $\pm$	3
			100	269 $\pm$	9
			200	378 $\pm$	4
			500	548 $\pm$	12
	Crude	20	0	56 $\pm$	1
			2	69 $\pm$	1
			4	104 $\pm$	5
			8	172 $\pm$	12
			10	142 $\pm$	7
			20	244 $\pm$	16
			50	325 $\pm$	37

(a) Underscore denotes activity greater than two times background

TABLE B2. (continued)

Boiling Point Cut	Fraction	S9 (μ l)	μ g/Plate	Revertant Colonies TA98/ μ g	
H-Coal 650-700°F (5226-094)	A ₁	100	0	64 $\pm$	3
			20	61 $\pm$	2
			40	69 $\pm$	2
			80	68 $\pm$	2
			100	54 $\pm$	2
			200	43 $\pm$	3
			500	49 $\pm$	1
	A ₂	100	0	64 $\pm$	3
			20	49 $\pm$	2
			40	58 $\pm$	1
			80	60 $\pm$	3
			100	44 $\pm$	1
			200	45 $\pm$	4
			500	52 $\pm$	2
	A ₃	100	0	64 $\pm$	3
			0.2	63 $\pm$	5
			0.4	102 $\pm$	8
			0.8	<u>121 $\pm$</u>	6 (a)
			1.0	<u>166 $\pm$</u>	4
			2.0	<u>274 $\pm$</u>	7
			5.0	<u>374 $\pm$</u>	10
	A ₄	100	0	64 $\pm$	3
			20	60 $\pm$	9
			40	60 $\pm$	3
			80	65 $\pm$	1
			100	57 $\pm$	4
			200	63 $\pm$	5
			500	53 $\pm$	2
	Crude	20	0	55 $\pm$	1
			2	49 $\pm$	0
			4	44 $\pm$	2
			8	56 $\pm$	2
			10	49 $\pm$	7
			20	48 $\pm$	5
			50	50 $\pm$	0

(a) Underscore denotes activity greater than two times background

TABLE B2. (continued)

Boiling Point Cut	Fraction	S9 (μ l)	μ g/Plate	Revertant Colonies TA98/ μ g	
H-Coal 700-750°F (5226-110)	A ₁	100	0	45 $\pm$	2
			20	42 $\pm$	4
			40	60 $\pm$	2
			80	49 $\pm$	6
			100	39 $\pm$	5
			200	45 $\pm$	2
			500	41 $\pm$	2
	A ₂	100	0	45 $\pm$	2
			20	70 $\pm$	2
			40	79 $\pm$	0
			80	65 $\pm$	2
			100	61 $\pm$	1
			200	59 $\pm$	2
			500	62 $\pm$	3
	A ₃	20	0	519 $\pm$	15
			0.2	518 $\pm$	8
			0.4	507 $\pm$	7
			0.8	657 $\pm$	39
			1.0	662 $\pm$	9
			2.0	999 $\pm$	11
			5.0	<u>1503 $\pm$</u>	<u>8</u> ^(a)
	A ₄	100	0	45 $\pm$	2
			20	86 $\pm$	3
			40	57 $\pm$	0
			80	81 $\pm$	3
			100	56 $\pm$	5
			200	65 $\pm$	1
			500	48 $\pm$	2
	Crude	20	0	39 $\pm$	4
			0.2	<u>81 $\pm$</u>	<u>6</u>
			0.4	<u>85 $\pm$</u>	<u>1</u>
			0.8	<u>92 $\pm$</u>	<u>5</u>
			1.0	<u>122 $\pm$</u>	<u>5</u>
			2.0	<u>120 $\pm$</u>	<u>9</u>
			5.0	<u>113 $\pm$</u>	<u>2</u>

(a) Underscore denotes activity greater than two times background

TABLE B2. (continued)

Boiling Point Cut	Fraction	S9 (μl)	μg/Plate	Revertant Colonies TA98/μg	
H-Coal 750-800°F (5226-111)	A ₁	100	0	28 ±	2
			20	41 ±	0
			40	49 ±	1
			80	36 ±	1
			100	40 ±	6
			200	45 ±	0
			500	39 ±	1
	A ₂	100	0	28 ±	2
			20	46 ±	3
			40	64 ±	1
			80	55 ±	2
			100	45 ±	4
			200	62 ±	4
			500	81 ±	1
	A ₃	20	0	519 ±	15
			0.2	485 ±	11
			0.4	577 ±	1
			0.8	602 ±	13
			1.0	705 ±	34
			2.0	911 ±	3
			5.0	<u>1397 ±</u>	<u>44</u> ^(a)
	A ₄	100	0	45 ±	1
			20	43 ±	7
			40	59 ±	4
			80	68 ±	3
			100	88 ±	3
			200	<u>106 ±</u>	<u>2</u>
			500	<u>115 ±</u>	<u>13</u>
	Crude	20	0	46 ±	1
			0.1	52 ±	6
			0.2	50 ±	3
			0.4	61 ±	1
			0.5	67 ±	1
			1.0	<u>107 ±</u>	<u>1</u>
			2.5	<u>155 ±</u>	<u>19</u>

(a) Underscore denotes activity greater than two times background

TABLE B2. (continued)

Boiling Point Cut	Fraction	S9 (μ l)	μ g/Plate	Revertant Colonies TA98/ μ g	
H-Coal 800-850°F (5226-112)	A ₁	100	0	55 $\pm$	5
			20	60 $\pm$	3
			40	35 $\pm$	2
			80	49 $\pm$	3
			100	47 $\pm$	6
			200	53 $\pm$	5
			500	50 $\pm$	6
	A ₂	100	0	55 $\pm$	5
			20	65 $\pm$	4
			40	82 $\pm$	1
			80	83 $\pm$	0
			100	80 $\pm$	10
			200	72 $\pm$	5
			500	56 $\pm$	4
	A ₃	20	0	519 $\pm$	15
			0.2	481 $\pm$	24
			0.4	560 $\pm$	13
			0.8	693 $\pm$	25
			1.0	956 $\pm$	6
			2.0	<u>1528 $\pm$</u>	<u>25</u>
			5.0	<u>2096 $\pm$</u>	<u>71</u>
	A ₄	100	0	55 $\pm$	5
			20	<u>198 $\pm$</u>	<u>9</u>
			40	<u>183 $\pm$</u>	<u>1</u>
			80	<u>239 $\pm$</u>	<u>12</u>
			100	<u>259 $\pm$</u>	<u>26</u>
			200	<u>364 $\pm$</u>	<u>23</u>
			500	<u>377 $\pm$</u>	<u>14</u>
	Crude	20	0	45 $\pm$	1
			0.2	60 $\pm$	9
			0.4	<u>91 $\pm$</u>	<u>5</u>
			0.8	<u>151 $\pm$</u>	<u>7</u>
			1.0	<u>133 $\pm$</u>	<u>9</u>
			2.0	<u>294 $\pm$</u>	<u>37</u>
			5.0	<u>481 $\pm$</u>	<u>10</u>

(a) Underscore denotes activity greater than two times background

TABLE B2. (continued)

Boiling Point Cut	Fraction	S9 (μ l)	μ g/Plate	Revertant Colonies TA98/ μ g	
H-Coal 850°+F (5226-109)	A ₁	100	0	30 $\pm$	1
			20	39 $\pm$	2
			40	55 $\pm$	1
			80	29 $\pm$	1
			100	43 $\pm$	5
			200	42 $\pm$	4
			500	38 $\pm$	1
	A ₂	100	0	30 $\pm$	1
			20	76 $\pm$	2 ^(a)
			40	95 $\pm$	1
			80	106 $\pm$	2
			100	112 $\pm$	9
			200	153 $\pm$	5
			500	201 $\pm$	3
	A ₃	20	0	44 $\pm$	1
			0.2	35 $\pm$	2
			0.4	58 $\pm$	5
			0.8	85 $\pm$	4
			1.0	163 $\pm$	10
			2.0	234 $\pm$	18
			5.0	728 $\pm$	22
	A ₄	100	0	446 $\pm$	3
			20	1153 $\pm$	14
			40	1609 $\pm$	18
			80	1877 $\pm$	46
			100	1510 $\pm$	17
			200	2111 $\pm$	51
			500	2339 $\pm$	16
	Crude	20	0	44 $\pm$	1
			0.2	43 $\pm$	2
			0.4	40 $\pm$	4
			0.8	76 $\pm$	3
			1.0	109 $\pm$	4
			2.0	210 $\pm$	2
			5.0	420 $\pm$	51

(a) Underscore denotes activity greater than two times background

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DISTILLATES AND H-COAL BLENDED MATERIAL

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