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# HYDROGEN BONDING IN ASPHALTENES AND COAL LIQUIDS

Quarterly Report for November 1, 1980—January 31, 1981

By
N. C. Li
L. Jones
N. F. Yaggi

Work Performed Under Contract No. AC22-80PC30252

Duquesne University
Department of Chemistry
Pittsburgh, Pennsylvania

**U. S. DEPARTMENT OF ENERGY**

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# HYDROGEN BONDING IN ASPHALTENES AND COAL LIQUIDS

Quarterly Report for the period
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N.C. Li, L. Jones and N.F. Yaggi
Department of Chemistry, Duquesne University
Pittsburgh, PA 15219

Prepared for
The U.S. Department of Energy
Under Contract No. DE-AC 22-80 PC 30252

## Objective and Scope of Work

The objective of this project is to investigate the nature and strength of hydrogen bonding and other molecular interactions in coal liquids and their fractions. Determinations are made of the molecular interactions involving the preasphaltene, asphaltene and oil, together with their separated fractions, obtained from coal liquids after hydroprocessing under different processing conditions and accelerated aging. NMR, IR, GC/MS, calorimetric, GPC, ESR, viscosity methods are used to determine structural changes caused by upgrading and aging of coal liquids.

Contract DE-AC 22-80 PC 30252 on Hydrogen Bonding in Asphaltenes and Coal Liquids has been awarded to Duquesne University for the period Aug. 1980-July, 1983. This is the second quarterly report, and describes the work accomplished from Nov. 1, 1980 to Jan. 31, 1981. During this period, a manuscript has been completed, with the title: Aging of SRC Liquids, by T. Hara, L. Jones, N.C. Li, K.C. Tewari. The paper is as follows:

ABSTRACT: The aging characteristics of 30/70 (wt. %) blend of SRC I and SRC II and of a SRC II Middle Distillate (boiling point range 450-565K), have been studied. Viscometric, electron spin resonance, ultimate analysis, solvent separations, gel permeation chromatography, and 600-MHz measurements have been used to monitor the aging properties of the SRC liquids under various conditions. The viscosity of the blend increases significantly in ten days with oxygen bubbling at $335^{o}K$. However, copper, in addition to oxygen, is necessary to significantly change the viscosity of the Middle Distillate. For the blend, there is a linear increase in the logarithm of viscosity with increasing wt. % of toluene-insolubles formed during oxygen-aging. There is also a linear decrease in the logarithm of viscosity with decreasing content of toluene-insolubles resulting from hydroprocessing of the blend with Ni-Mo catalyst. These observations indicate that under conditions of oxidative degradation the formation and content of larger molecular-size toluene-insolubles are mainly responsible for the increased viscosity of the blend. The middle distillate does not contain toluene-insolubles, and oxygen-aging occurs only in the presence of copper. Oxidative coupling of phenols is proposed as an aging mechanism for both the blend and the middle distillate.

Recent technological development has demonstrated that coal-derived liquids can be used as a low-sulfur boiler fuel as well as a feedstock for further refining to produce a more acceptable fuel for home, transportation and industrial use. There exists a need to determine storage characteristics which affect the handling properties and burning efficiency of coal-derived liquids. However, very little information is available on the mechanism of fuel deterioration in natural and accelerated aging conditions.[1-6]

One of the major influences upon aging is that of oxidation. Several investigators[4-6] have found that coal-derived liquids are very susceptible to oxidative degradation, and that oxygen exerts a dramatic effect on enhancing the viscosity of coal-liquid fuels.

In the present study, two different kinds of solvent-refined coal (SRC) liquids were subjected to aging in order to clarify a reaction mechanism involved in the oxidative degradation of coal liquids. A 30/70 blend of solid product (SRC I) with liquid product (SRC II), as well as a middle distillate of SRC II, were chosen for the aging study. The blend is a typical heavy coal liquid containing a considerable amount of toluene-insolubles and asphaltene, and has been used as a feedstock for catalytic hydroprocessing. The middle distillate is entirely composed of pentane-soluble oil and can be used as a low-sulfur boiler fuel.

## EXPERIMENTAL

The SRC I and SRC II, used for the preparation of the blend, were made from western Kentucky bituminous coal at the Fort Lewis SRC Pilot Plant. The blend was prepared by gradual addition of 30 parts by weight of powdered SRC I to 70 parts by weight of SRC II under nitrogen with constant stirring for 2.5 h at 413-423K. SRC II-Middle Distillate (boiling point range 450-565K) was made from Pittsburgh seam coal and was obtained from a process run operated at 13.8 MPa hydrogen pressure, 730K and 1 h residence time. Prior to aging runs the liquids were stored under nitrogen at 273K.

Accelerated aging of the blend and SRC II - Middle Distillate was carried out in a three-necked flask (200 ml) equipped with a gas bubbling inlet and a condenser. The inlet tube passed nearly to the bottom of the flask allowing oxygen (or nitrogen) to be bubbled through the coal liquid at a flow rate of about 2 ml/min. A constant temperature bath was used to maintain the temperature at 335K. The accelerated aging under oxygen of SRC II - Middle Distillate was carried out in the presence of copper shavings (2 wt % of the coal liquid).

The solvent separation of the blend, before and after aging, into oil (pentane- and toluene-soluble), asphaltene (toluene soluble) and toluene-insoluble fractions was accomplished using a scheme described elsewhere[7]. The pentane- and toluene-soluble oil was further fractionated into acidic, basic, aromatic and saturate fractions by sequential treatment with anion-exchange resin (Amberlite IRA-904), cation-exchange resin (Amberlite A-15) and silica gel. The details of the method have been described elsewhere[8,9]

The viscosities of the liquids were determined at 303K or 333K using a Brookfield viscometer equipped with a small sample adaptor. Infrared spectra were recorded on thin film between NaCl plates or as KBr pellets with a Beckman IR-20 spectrometer. Solution infrared spectra were recorded from dilute solutions in carbon disulfide in a 5-mm KBr liquid cell with the solvent in the compensating beams. Proton magnetic resonance spectra were obtained on $CDCl_3$ solutions with a 600-MHz NMR spectrometer at Carnegie-Mellon University. Tetramethylsilane was used for the lock signal as well as the internal reference. Electron spin resonance (ESR) spectra of toluene-insoluble fractions were obtained on a Varian E-4 spectrometer equipped with E-204 low frequency modulation module. ESR parameters were determined according to method described elsewhere[10-12]. The toluene-insoluble fractions were dried under vacuum at 363K for 48 h, weighed in a quartz ESR tube and sealed after evacuation at $10^{-2}$ PA with heating for 2 h. A capillary containing an aqueous solution of Fremy's salt was used to calibrate the magnetic field. Spin concentration in the toluene-insoluble fraction isolated from the aged blend relative to that derived from unaged blend was determined using double integration of the signals obtained under the same operational conditions.

Molecular size distributions was obtained by gel permeation chromatography (GPC). The two sets of columns used were: (a) $10^4$, $10^3$, 500 nm μ-styragel and two 50 nm μ-sherogel, and (b) Shodex A-80M and A-802. The instrument was operated at ambient temperature with tetrahydrofuran (THF) as the mobile phase at a flow rate of 2.0 or 1.0 ml/min.

## RESULTS AND DISCUSSIONS

The results of solvent fractionation, elemental analysis, specific gravity and viscosity of the unaged 30/70 blend of SRC I/SRC II and SRC II-Middle Distillate are listed in Table 1. Storage of the blend under nitrogen at about 273K for 120 days showed no significant change in the properties listed in Table 1.

Figure 1 shows the changes in viscosity that occur when the blend was stored at 335K, both in dark and light, and subjected to oxygen and nitrogen bubbling. It can be seen that exposure to oxygen had the most pronounced effect upon the increase in viscosity of the blend. Exposure to nitrogen and light has a small effect in increasing the rate of degradation of the blend. Over a period of two days, aging with nitrogen bubbling at 335K increased the viscosity of the blend from 36 cp to 70.4 cp. Although the vessel was equipped with a condenser, part of the viscosity increase may well be due to the loss of the volatile components under constant gas bubbling.

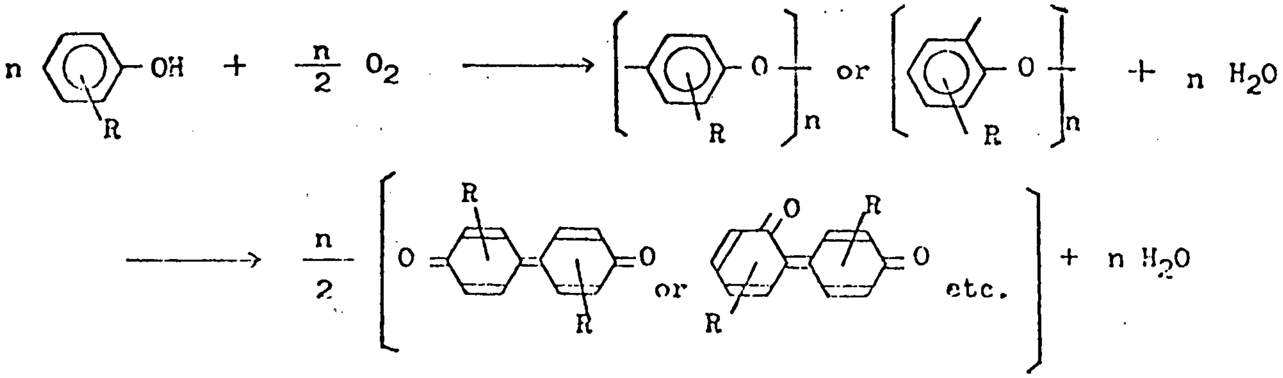
During oxygen aging of the blend, aliquots of the coal liquid were removed from the container at various time intervals and were solvent fractionated. Figure 2 shows the weight percent of the different fractions isolated as a function of the aging time. The content of toluene-insolubles increased significantly with the aging time, accompanied by a corresponding decrease of asphaltene and oil fractions. From Figure 2, it is also obvious that the asphaltene components contribute more to the increase in content of toluene-insolubles than the pentane-soluble oil fraction. The rectilinear dependence of logarithm of viscosity ( $\ln \eta$) of the blend under oxygen aging with the content of toluene-insolubles is shown in Figure 3. It is interesting to note that the rectilinear dependence of $\ln \eta$ with content of toluene-insolubles is also observed when the same blend was hydroprocessed over Ni-Mo catalyst[13]. This is also illustrated in Figure 3. The above observations indicate that during oxidative degradation the formation and content of larger molecular-size toluene-insolubles are mainly responsible for the increased viscosity of the blend.

Infrared spectra of the isolated fractions from both aged and unaged blend have been obtained. After accelerated aging with oxygen for 10 days, both the toluene-insoluble and asphaltene fractions show an additional absorption band at about 1700 $cm^{-1}$ characteristic of a carbonyl group. This absorption does not appear in oxygen-aged oil fractions. Brown and Karn[5] have reported that the oxygen content of a whole sample increased from 2.2 (unaged) to 3.1 wt % (oxygen-aged), which shows that oxygen has been chemically incorporated into the sample. A band at about 1700 $cm^{-1}$ is present only for oxygen-aged sample, but not for unaged or nitrogen-aged sample. The oxygen content in the asphaltene and benzene-insoluble fractions increased with oxygen-aging, whereas a decrease was found in the oil fraction. Our infrared results lend support to the suggestion that oxygen is incorporated preferentially into the asphaltene and toluene-insoluble fractions, resulting in formation of carbonyl groups in these higher-molecular weight fractions.

GPC profiles of the blend and its fractions, before and after oxygen aging, are shown in Figure 4. It is clear that on aging, the formation of larger-size components predominantly occurs for the toluene-insoluble fractions. The profiles of pentane-soluble oil fractions do not show the formation of larger particles, but the decrease of the peak at elution volume of 48 ml is noticeable after aging for 10 days.

Infrared spectrum of the aged oil fraction in dilute $CS_2$ solution showed marked decrease of free phenolic OH stretching at 3600 $cm^{-1}$ relative to that of unaged oil fraction. The results suggest that in the oil fraction the phenolic components preferentially react to form larger molecules which are substantially insoluble in pentane.

Oxidative coupling of phenols has been well studied[14] and proceeds through the following mechanism:



Scheme I

The reaction products are C-O polyphenylene ethers of high molecular weights and C-C coupling products of diphenoquinones. The selective formation of a particular product depends on the catalyst, the solvent, and the phenolic compound used. A polymerization reaction is more likely than dimerization in many cases[15]. Reactivity of oxidative coupling of various phenolic compounds depends upon the oxidation potentials of phenols, since the rate-determining step exists in the electron-transfer process from a phenoxy anion to a phenoxy radical[14]. The oxidation potentials among dihydroxy and monohydroxybenzenes increase in the following order: hydroquinones < pyrocatechols < resorcinols < monohydroxybenzenes. Consequently, rates of oxidation decrease in the same order[14]. In the aging of the blend with oxygen, the formation of larger molecules occurs predominantly in fractions composed of higher molecular-size components. In other words, the rate of oxidative degradation may be varied in the following order: toluene-insolubles > asphaltenes > oil. Since the organic structure of coal contains dihyroxybenzene units as a quinizarin structure[16,17], (1,4-dihydroxyanthraquinone) it may be possible to consider that fractions composed of higher molecular-size components contain large amounts of dihydroxybenzene derivatives, resulting in higher reactivity to oxidative coupling. The weak absorption at 1700 $cm^{-1}$ in the infrared spectra of aged toluene-insoluble and asphaltene fractions can be considered as due to the formation of dimers as minor reaction products.

Table 2 lists the changes in ESR parameters of the toluene-insoluble fraction of the blend during aging at 335K. The observed decrease in spin concentration of toluene-insoluble fraction with oxygen aging is probably not due to radical-radical combination. Figure 2 shows that part of the aged toluene-insoluble fraction is formed from the asphaltene and oil fractions of the blend, and Goldberg et al.[12] have shown that the spin concentrations in the fractions decrease drastically in the order preasphaltene (toluene-insoluble) > asphaltene > oil. A small increase in g-value with aging time may well be due to the contribution of semiquinone or phenoxy radicals formed by the reaction of di- or mono- phenols with oxygen[10]. Aging with nitrogen shows only a small effect on ESR parameters.

The accelerated aging of SRC II Middle Distillate in the presence of copper shavings also supports the above degradation mechanism. Copper(II) complexes have been widely used as effective catalysts for oxidative coupling of monophenols[14,15]. Besides, strips of copper foil[18] and some copper salts such as chloride[19] or naphthenate[3] are found to have an accelerating effect on the aging of coal-derived materials and heavy ends of petroleum products.

The viscosity of SRC II-Middle Distillate did not significantly increase by bubbling with oxygen at 335K for 5 days (Figure 5). The large molecular-size toluene-insoluble fraction, which is mainly responsible for the viscosity of the SRC I-SRC II blend, is absent here. However, if oxygen is bubbled through the Middle Distillate in the presence of copper, viscosity increases drastically, similar to the blend. The presence of copper alone, without oxygen, does not increase the viscosity. GPC profiles of the aged Middle Distillate clearly show the formation of larger molecular-size compounds by the reaction with oxygen and copper (Figure 6). On aging of the Middle Distillate, solution infrared spectra show a significant decrease in the intensity of the free phenolic OH stretching vibration at 3600 $cm^{-1}$. NMR spectra in the region of aromatic protons are shown in Figure 7. After aging the Middle Distillate with oxygen and copper at 335K for 4 days, the aged liquid was separated into pentane-soluble oil and pentane-insoluble precipitate by extraction with 20-fold excess pentane. The NMR spectrum of the pentane-soluble oil fraction of the aged Middle Distillate is shown in Figure 7. The spectrum shows that aromatic protons whose signals appear in higher magnetic field (6.40-6.75 ppm) disappear or are reduced significantly when compared with the unaged liquid. It is interesting to note that this aspect of the NMR spectra of unaged and aged pentane-soluble Middles Distillate is quite similar to that of the spectra of the oil fraction of the blend and its chromatographically separated acid-free subfraction, also shown in Figure 7. It is well known that aromatic proton signals show upfield shift when electron-donating groups such as hydroxyl are attached to the same ring structure. In conclusion, both IR and NMR results indicate that acidic components (mainly phenols) are selectively reacted in the accelerated aging of the SRC II Middle Distillate to form higher molecular-weight components, which are responsible for the increase in viscosity of the coal-derived liquid. For the blend, we have proposed oxidative coupling of phenols as an aging mechanism. The accelerated aging of the Middle Distillate, in the presence of copper, also supports the proposed mechanism for oxidative degradation.

According to the mechanism of oxidative coupling of phenols (Scheme I), for R = $CH_3$, the initial atomic ratio $H/C = 8/7 = 1.1$. After reaction with oxygen, H/C becomes $(8-2)/7 = 0.9$. We have analyzed the pentane-insoluble precipitate of the Middle Distillate, obtained after reaction with oxygen and copper for 5 days at 335K, and found that $H/C = 1.0$, as compared to $H/C \approx 1.2$ for the unaged Middle Distillate. Comparison of the results of carbon and hydrogen analysis for the unaged Middle Distillate and for the pentane-insoluble precipitate obtained after aging, therefore, lends further support to Scheme I for oxidative degradation.

## ACKNOWLEDGMENT

We acknowledge the support of the U.S. Department of Energy under Contract No. DE-AC 22-80PC 30252. The 600-MHz NMR spectrometer at Carnegie-Mellon University, supported by PHS Grant No. RR-00292, was used for obtaining the NMR spectra. We thank David Pratt for use of the ESR spectrometer at the University of Pittsburgh and Laurine Galya for obtaining the GPC chromatograms of Figure 4. We are grateful to B.C. Bockrath and D. Finseth for helpful discussions.

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TABLE 1 PROPERTIES OF TWO SRC LIQUIDS FOR AGING STUDY

| Coal Liquid | | 30-70 blend of SRC I with SRC II | SRC II MIDDLE DISTILLATE |
|---|---|---|---|
| | C | 86.2 | 86.05 |
| ELEMENTAL | H | 7.85 | 8.95 |
| ANALYSIS | O | 4.2 | 3.8 |
| | N | 1.32 | 1.04 |
| (wt % maf) | S | 0.45 | 0.17 |
| ATOMIC RATIO OF H/C | | 1.08 | 1.24 |
| SPECIFIC GRAVITY (289 K) | | 1.07 | 0.974 |
| VISCOSITY | | 36 | 5.0 |
| (cP) | | at 333 K | at 303 K |
| SOLVENT FRACTIONATION | | WT% | WT% |
| TOLUENE INSOLUBLE | | 10.2 | 0.0 |
| ASPHALTENE | | 18.5 | 0.0 |
| PENTANE-SOLUBLE OIL | | 71.3 | 100.0 |

Table 2 Change in ESR parameters of toluene-insoluble fraction of SRC blend during aging with oxygen at 335K

| Sample | g-value | line width (gauss) | relative spin concentration |
|---|---|---|---|
| Before aging | 2.0031 | 6.2 | 1.00 |
| Aging with $O_2$ for 5 days | 2.0032 | 5.7 | 0.86 |
| Aging with $O_2$ for 8 days | 2.0033 | 5.3 | 0.68 |
| Aging with $O_2$ for 10 days | 2.0034 | 5.1 | 0.53 |
| Aging with $N_2$ for 10 days | 2.0032 | 6.2 | 0.89 |

Captions for figures

Figure 1 Viscosity changes of the SRC blend during aging.

Figure 2 Changes in compositions of the SRC blend during aging.

Figure 3 Plots of logarithm of viscosity with wt.% toluene-insolubles in (A) hydroprocessing and (B) aging experiments on SRC blend. Viscosity (A) 311K, (B) 333K.

Figure 4 GPC of SRC blend and its fractions before and after aging with oxygen at 335K for 10 days. Solid lines, before aging; dotted lines, after aging. GPC conditions: columns, $10^4$, $10^3$, 500 mn $\mu$-styragel + two 50 nm $\mu$-spherogel. Mobile phase: THF, 2.0 ml/min. RI detector.

Figure 5 Viscosity change of SRC II Middle Distillate during aging at 335K

(a) with copper and oxygen
(b) with oxygen alone
(c) with copper alone

Figure 6 Molecular-size distribution profile of SRC II

Middle Distillate before and after aging: Unaged ____, $O_2$-aged -----, $O_2$- and copper-aged ...... Columns: Shodex A-80M, A-802, Mobile phase: THF, 1 ml/min. UV 254 detector.

Figure 7 600-MHz NMR spectra A-1, unaged SRC II Middle Distillate; A-2, pentane-soluble fraction of aged SRC II Middle Distillate with oxygen and copper at 335K for 4 days; B-1, pentane-soluble oil of the blend; B-2, acid-free subfraction of B-1.

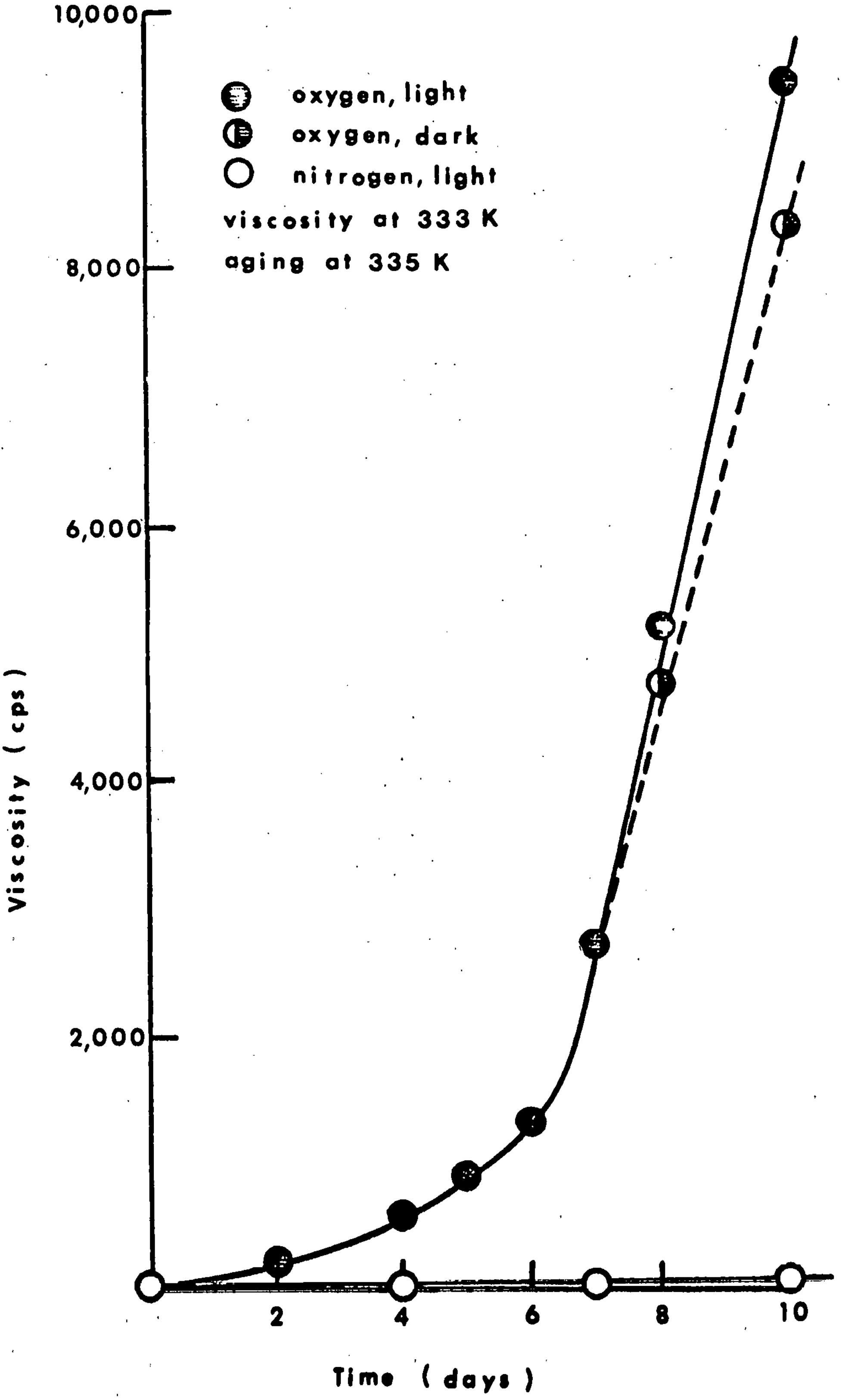

10,000
8,000
6,000
4,000
2,000
oxygen, light
oxygen, dark
nitrogen, light
viscosity at 333 K
aging at 335 K
Viscosity ( cps )
2
4
6
8
10
Time ( days )

Figure 1

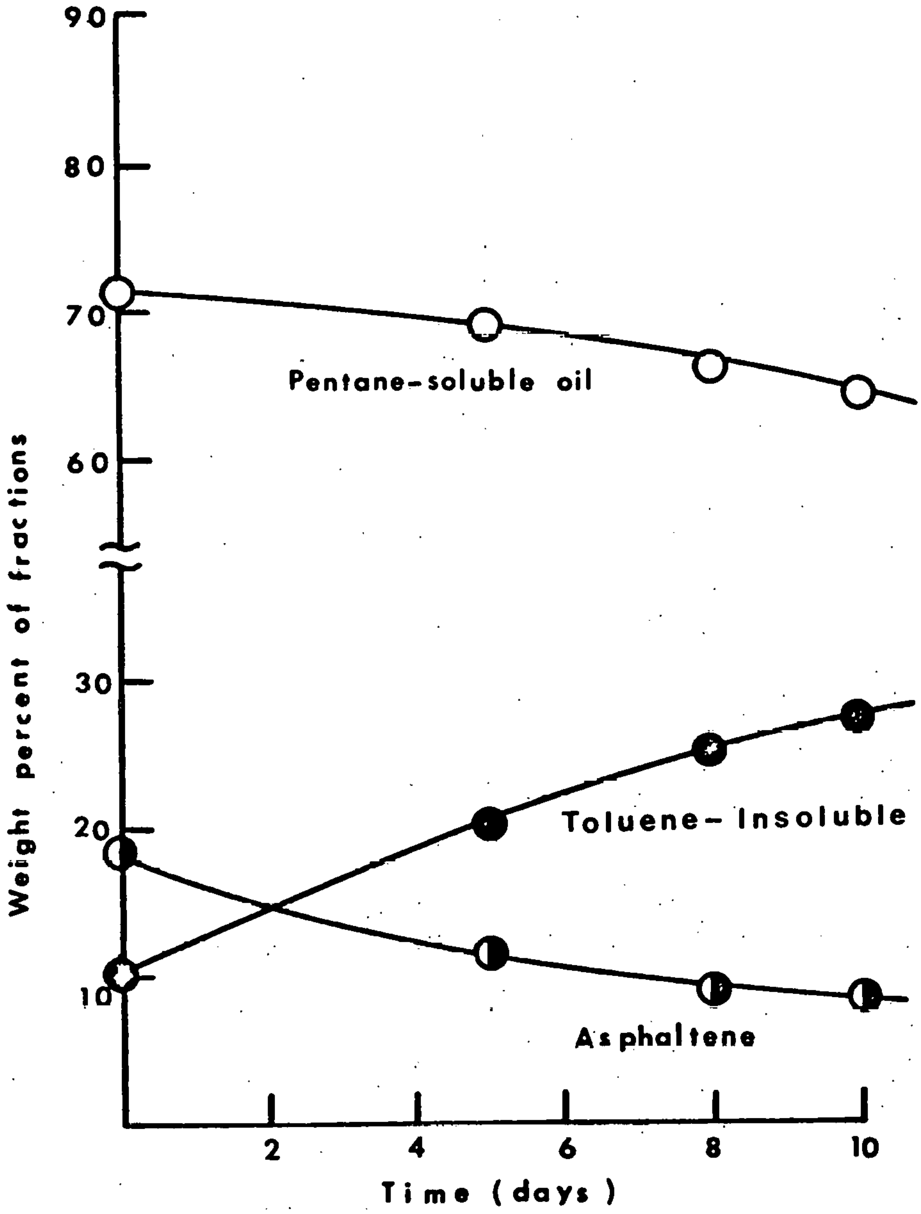
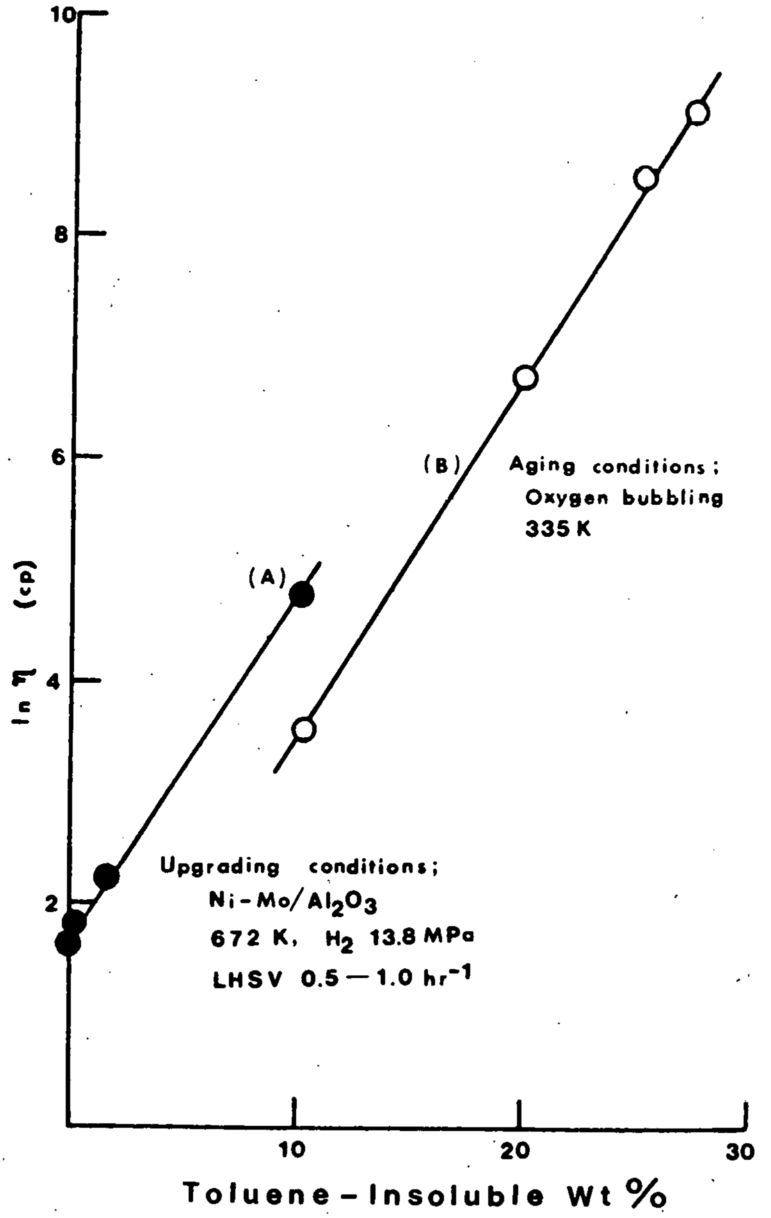

90
80
70
60
30
20
10
Weight percent of fractions
Pentane-soluble oil
Toluene-Insoluble
Asphaltene
2
4
6
8
10
Time (days)

Figure 2



10
8
6
4
2
ln η (cp)
(A)
(B)
Aging conditions;
Oxygen bubbling
335 K
Upgrading conditions;
Ni-Mo/Al$_2$O$_3$
672 K, H$_2$ 13.8 MPa
LHSV 0.5 — 1.0 hr$^{-1}$
10
20
30
Toluene-Insoluble Wt %

Figure 3

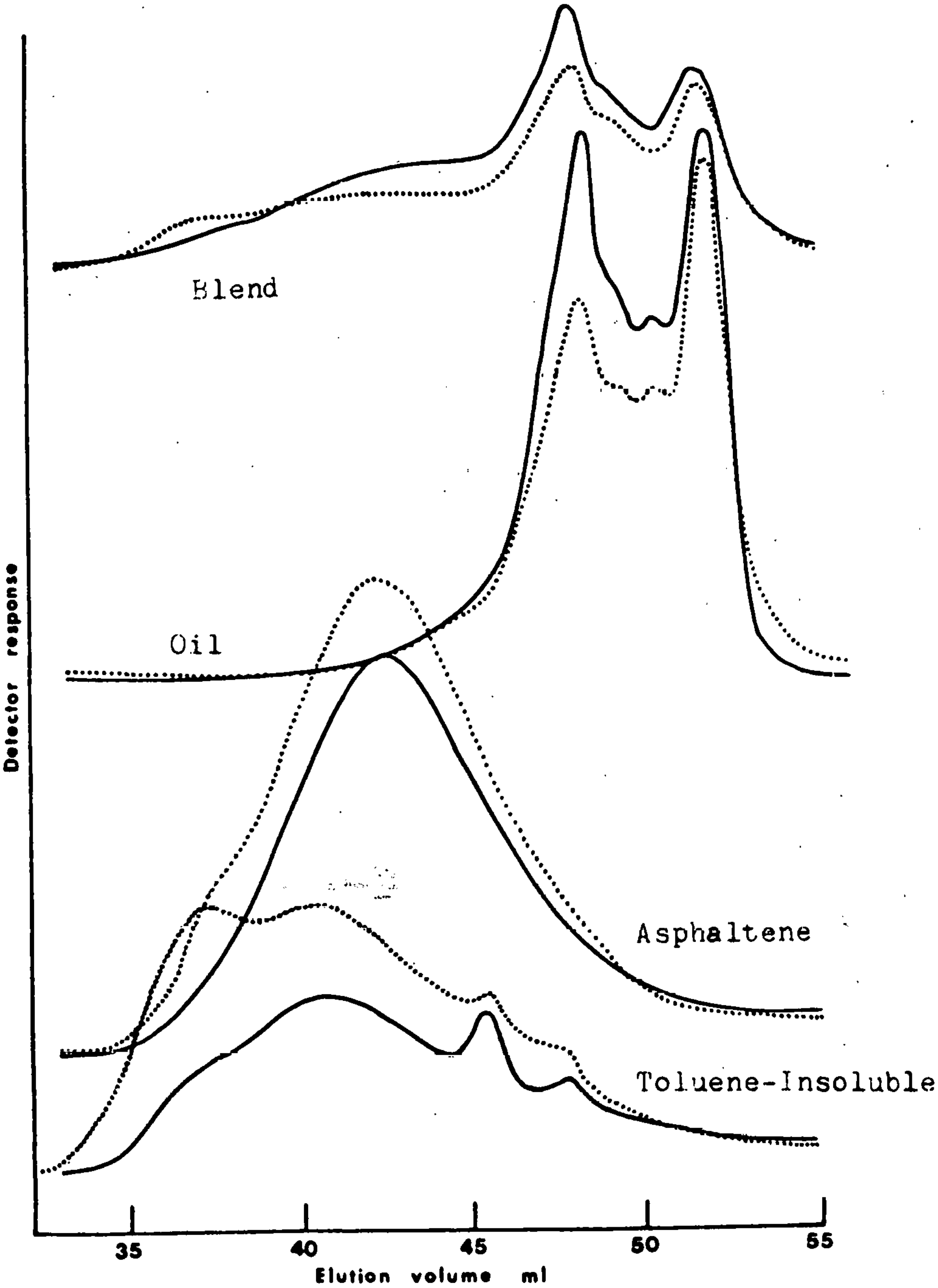

Blend
Oil
Asphaltene
Toluene-Insoluble
Detector response
35
40
45
50
55
Elution volume ml

Figure 4

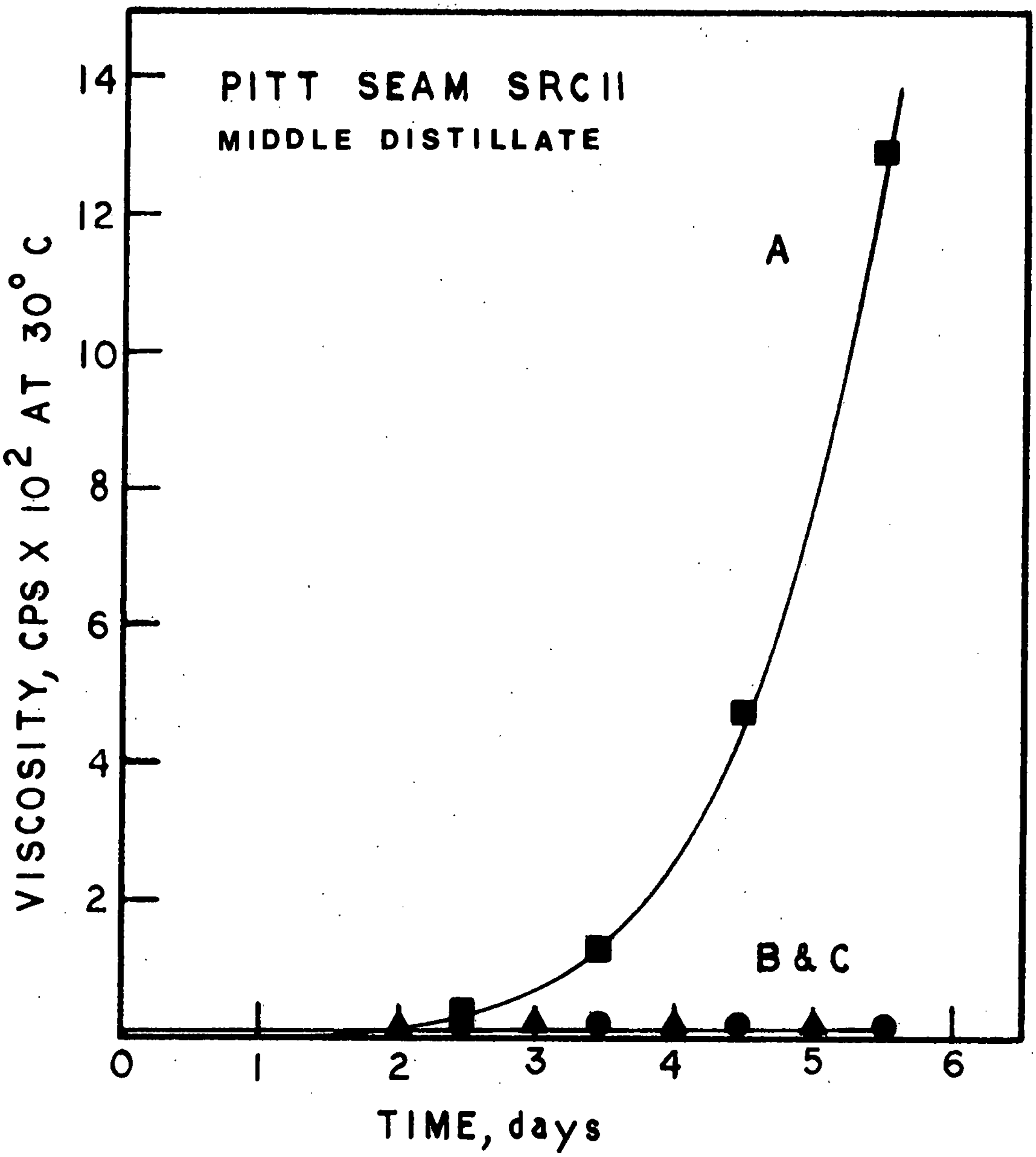

PITT SEAM SRC II
MIDDLE DISTILLATE
A
B & C
VISCOSITY, CPS X $10^2$ AT 30° C
14
12
10
8
6
4
2
0
1
2
3
4
5
6
TIME, days

Figure 5

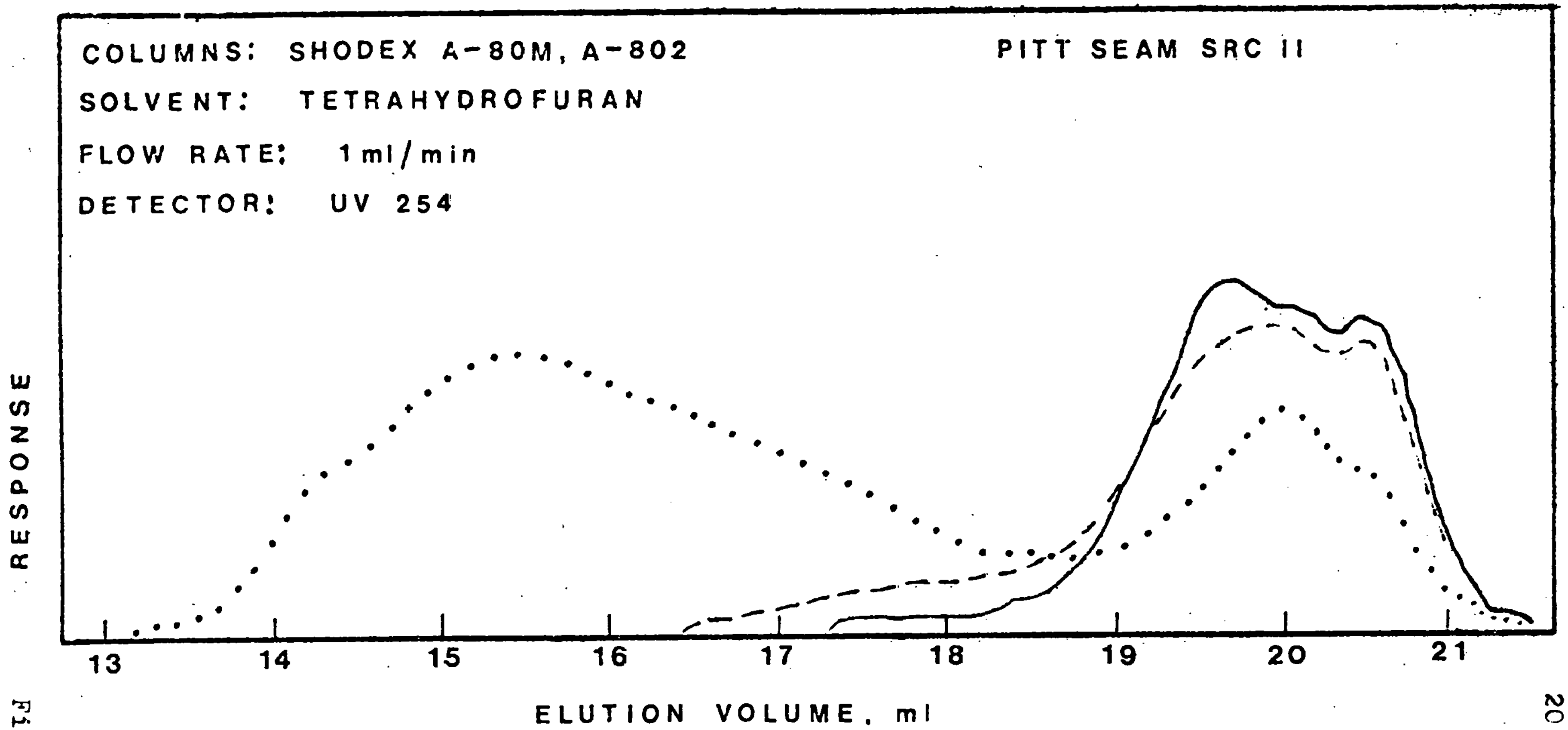

COLUMNS: SHODEX A-80M, A-802
SOLVENT: TETRAHYDROFURAN
FLOW RATE: 1 ml/min
DETECTOR: UV 254
PITT SEAM SRC II
RESPONSE
13
14
15
16
17
18
19
20
21
ELUTION VOLUME, ml

Figure 6

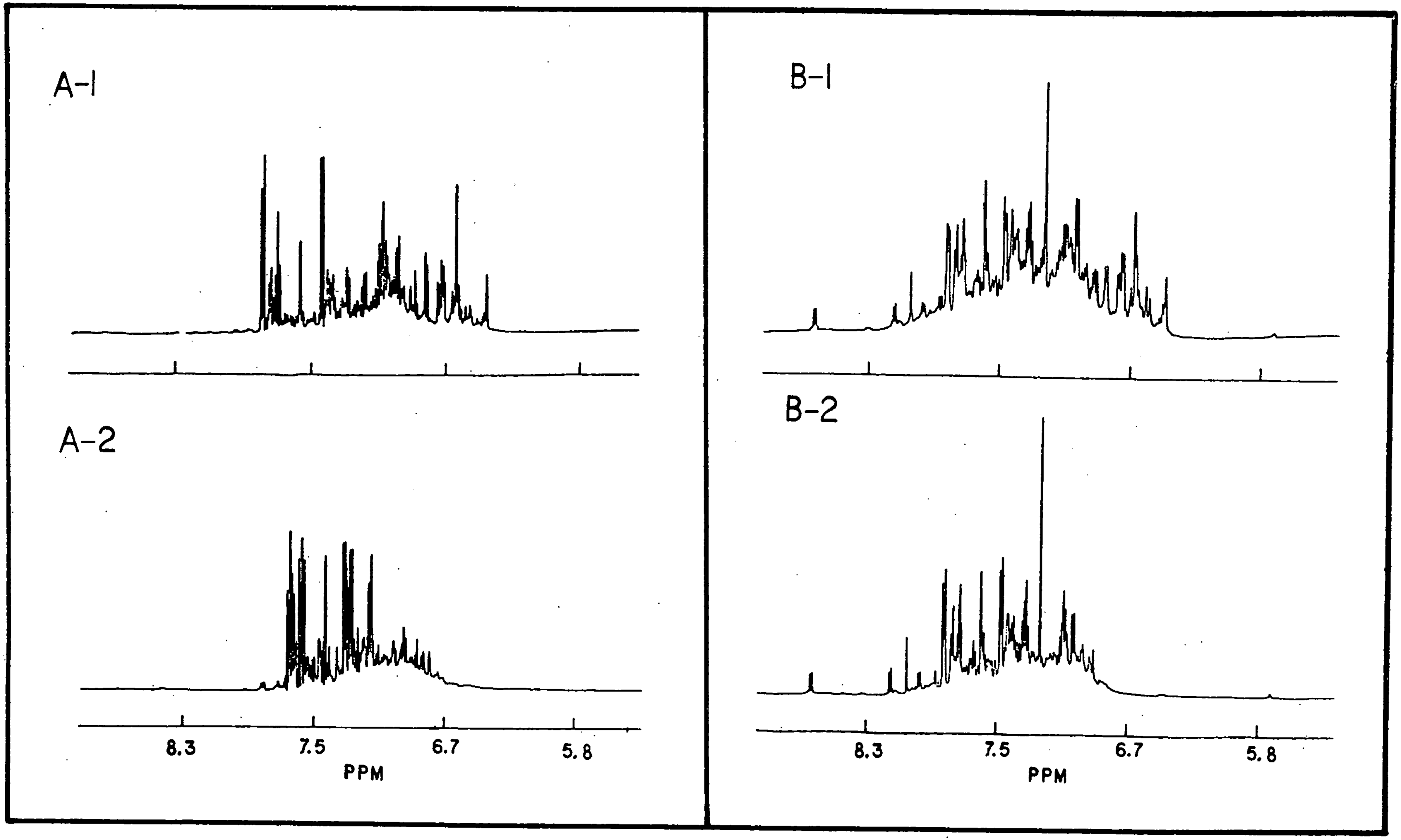


Figure 7

## Work in Progress on Aging Study of Coal-Derived Liquids

The completed manuscript deals with aging study of a 30/70 blend of SRC I/SRC II, and of a SRC II Middle Distillate made from Pittsburgh seam coal. Work is in progress on aging study of a 20/80 blend of SRC I/SRC II at 25°, 50°, 60° and 90°, and of a SRC II Middle Distillate made from Illinois No. 6 coal at 62°C, in the presence of copper. The effect of additives: phenol, anisole, o-phenylphenol and other phenol derivatives on the rates of aging is being investigated.

For SRC II Middle Distillate made from Illinois No. 6 coal, there is an exponential increase in viscosity with time when the coal liquid is aged at 62°C with oxygen and copper, similar to the curves shown in Figure 5 for the Middle Distillate made from Pittsburgh seam coal. While some loss of volatile components undoubtedly occurs which would contribute to an increase in viscosity of a given sample, the fact that copper alone or oxygen alone at 62° does not affect the viscosity in five days, must mean that the presence of both copper and oxygen is necessary in order to have an impact on the aging of Middle Distillates. If the Middle Distillate is aged at 62° with oxygen, without copper, it is necessary to wait 12 weeks before viscosity begins to increase. This indicates that aging occurs at a much slower rate, in the absence of copper.

The following techniques are being used: NMR (60-MHz and 600-MHz), Fourier-transform infrared, gel permeation chromatography, gas chromatography, HPLC and ultimate analysis of the pentane-soluble and pentane-insoluble fractions. The proposed mechanism of aging, i.e., oxidative coupling of phenols, is being subject to further tests.

PERSONNEL

During the period November 1, 1980 to Jan. 31, 1981 persons participating in the research are as follows: Dr. N.C. Li, 1/3 time, Dr. L. Jones, full-time, Dr. N.F. Yaggi, full-time. L.J.S. Young, J. Dru, J. Ge and R. Gross are students.

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