Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Ijam, Mousa J. and Arif, Saffa F.(1992) 'Identification of Nonhydrocarbon Aromatic Compounds in the Gas Oil Fraction of Kuwait Petroleum', Separation Science and Technology, 27: 1, 73 — 86
To link to this Article: DOI: 10.1080/01496399208018866
URL: http://dx.doi.org/10.1080/01496399208018866

SEPARATION SCIENCE AND TECHNOLOGY, **27**(1), pp. 73–86, 1992

Identification of Nonhydrocarbon Aromatic Compounds in the Gas Oil Fraction of Kuwait Petroleum

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Abstract

The amounts of individual components in the nonhydrocarbon aromatic portion of a Kuwait gas oil boiling in the range from 218 to 345°C are reported. Twenty-eight nonhydrocarbon aromatic components were isolated and identified from their separated fractions. The majority are C_1–C_5 benzo- and dibenzothiophenes. The compounds were isolated principally by using preparative scale high performance liquid chromatography, and identified by nuclear magnetic resonance and gas chromatography supplemented by mass spectrometry.

INTRODUCTION

For several decades the removal of aromatics from crude oil fractions (e.g., kerosene and lubricating oils) has been practiced in oil refining to produce fuels and lubricants of lower aromatic content and hence of improved quality. These aromatics are suitable raw materials for the manufacture of aromatic solvents, aromatic process oils, high octane gasoline, and as basic materials for making detergents, perfumes, and dyes. Specific methods which have been applied for aromatic removal are: a) oleum treatment, b) silica gel adsorption, c) hydrogenation, and d) solvent extraction. Much of the developmental work has utilized activated silica gel as the liquid chromatography stationary phase (*1–6*).

The composition of aromatic compounds has been investigated in detail by means of adsorption liquid chromatography on different sorbents (*7–11*). Monocyclic, bicyclic, and tricyclic aromatic compounds were separated, and the quantitative analyses of aromatics in each of these three classes were used for calculating the composition of the aromatics in their mixtures.

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A study of the UV and IR spectra of the aromatics showed them to consist mainly of bi-, tri-, tetra-, and penta-substituted benzene, bicyclic, and tricyclic aromatic compounds. Detection and identification have been also carried out by capillary gas chromatography (*12*) and by gas chromatography–mass spectrometry (GC/MS) (*13, 14*). Recently, high performance liquid chromatography (HPLC) which fractionates the aromatic compounds in petroleum into classes of compounds has received considerable attention (*15, 16*).

Detailed studies of molecular structure and substituent effects have been reported on the retention characteristics of aromatic compounds on alumina, silica, and various chemically bonded silicas containing $-C_{18}$, $-NH_2$, $-R(NH_2)_2$, —CN, RCN, and phenyl-mercuric acetate for compound class (ring-numbered) high performance liquid chromatography separation (*17–21*).

In previous work in this laboratory (*5, 6, 22, 23*) normal and branched aliphatic hydrocarbons were isolated and identified from kerosene and light gas oil. This paper describes the extension of this previous work to the separation and identification of aromatic ring classes (mono-, bi-, and tricyclic aromatics) in the gas oil fraction of Kuwait petroleum.

Identification of the major components in the nonhydrocarbon aromatics was our primary objective in this study.

EXPERIMENTAL

Apparatus

The instrument used for detection of nonhydrocarbon aromatics was a Varian model 60-A nuclear magnetic resonance (NMR) spectrometer.

Chromatographic separation of the gas oil (GO) into classes of compounds (mono-, bi-, and tricyclic aromatics) was performed via column liquid chromatographic fractionation followed by GC/MS. Analysis was performed by using a Finnigan model 6612-4000 fitted with a 30-m, fused silica capillary column, internally coated with a 0.25-mm layer of DB-5 stationary phase. The chromatography was programmed from 4°C/min to 310°C. A Grob split-splitless injector was used in the splitless mode and maintained at 280°C. Helium carrier gas was used at 7 psi and a flow rate of 1.2 mL/min. The column/spectrometer interface temperature was maintained at 300°C. MS parameters included: source temperature set at 270°C, electron impact ionization at 58 V, source pressure at 1.2×10^{-7} torr. The ionizer filament turned on 4 min after injection.

A Shimadzu model LC-4A high performance liquid chromatograph (HPLC) was used for separation and identification of the neutral and basic aromatic fraction. The instrument was equipped with UV (254 nm wavelength) and RI detectors, attached to a data processor C-R3A chromatopac (176 Kbyte) memory capacity, and connected with preparative stainless steel column (25 cm length, 2.12 cm i.d.) of ordinary phase SIL Shimpac and reverse phase C_{18}-ODS type.

The products were quantitated by a GC/MS Hewlett-Packard 5993 system. The equipment was provided with disc drive 7906 HP and recorder 1876 A, and connected to a fused silica capillary column of 2.5 cm length coated with OV-101. The measurements were carried out under the following conditions: Helium flow 1.2 mL/min, injection port 250°C, scan start delay 0.8 min, purge time 0.5 min, electron multiplier detector at 1600 V, temperature programming from 50°C at 10°C/min, rate of increase to 250°C for 3 min.

Reagents

Gas oil (bp 218–345°C, distallate fraction of crude oil) was obtained from Kuwait National Petroleum Company (KNPC). Its physical properties are summarized in Table 1. Commercially available HPLC grade solvents were used without further purification (the gas chromatograms of the solvents were obtained, no extra peaks), e.g., *n*-pentane (product of Pierce Chemical Company, Rockford, Illinois 61105, USA), *n*-hexane (product

TABLE 1
Physical Properties of the Gas Oil Sample

Characteristics	ASTM method	Result
Specific gravity, δ 15.6/15.6°C	D1298-80	0.8343
API gravity, δ 15.6°C		38.1
Density, δ 15°C, g/cm^3		0.8340
Flash point, °C	D93-80	92.2
Aniline point, °C	D611-82	71.6
Diesel index		59.7
Refractive index at 20°C, n_D^{20}	D1747-62	1.46539
Cloud point, °C	D2500-66	−9
Pour point, °C	D97-66	−12
Kinematic viscosity at 40°C	D445-79	3.10
Kinematic viscosity at 100°C		1.18
Aromatics, %	D1319-77	22.0
Olefins, %		0.8
Paraffins + naphthenes, %		77.2
Color	D1500-64	0.5
Ash content (on 10% res) Wt%	D484-80	0.008
Boiling point, °C	D86-78	218–345

of J. T. Baker Chemical Co., Phillipsburg, New Jersey 08865, USA), methanol, ethanol, tetrahydrofuran, and methylene chloride (products of Applied Science Labs., Alltech Associates, Inc., 2051 Waukegan Road, Deerfield, Illinois, USA).

Chromatographic Procedure

Chromatographic separation was performed using a special apparatus designed and constructed locally (Fig. 1). In a typical experiment, the gas oil (100 g) was chromatographed on activated silica gel (450 g, 100–140 mesh) from solution in pentane (bp 28°C, 300 mL) at a temperature of 15°C. The gel was the product of Eschwege Woelm Pharma, Germany. The gel was dried before use in an oven at 250°F in a shallow vessel for 5 h, and the dried gel was transferred to an air-tight container while still hot.

The column was eluted with 120 mL pentane. Paraffins were collected in 12 fractions of 100 cm^3 each. The elution was stopped at the first appearance of aromatics in the NMR spectra of the last fractions. Distillation

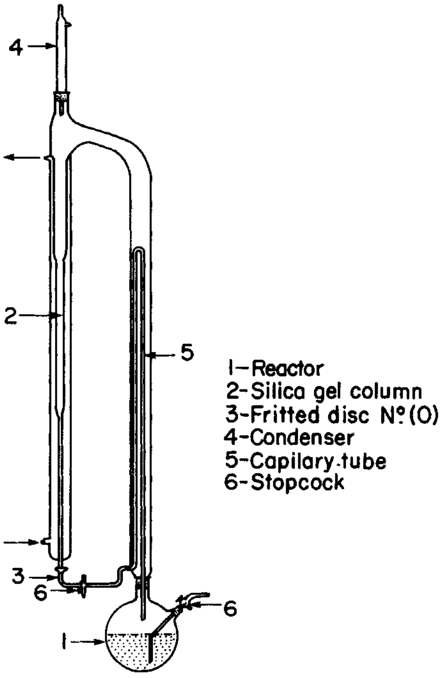
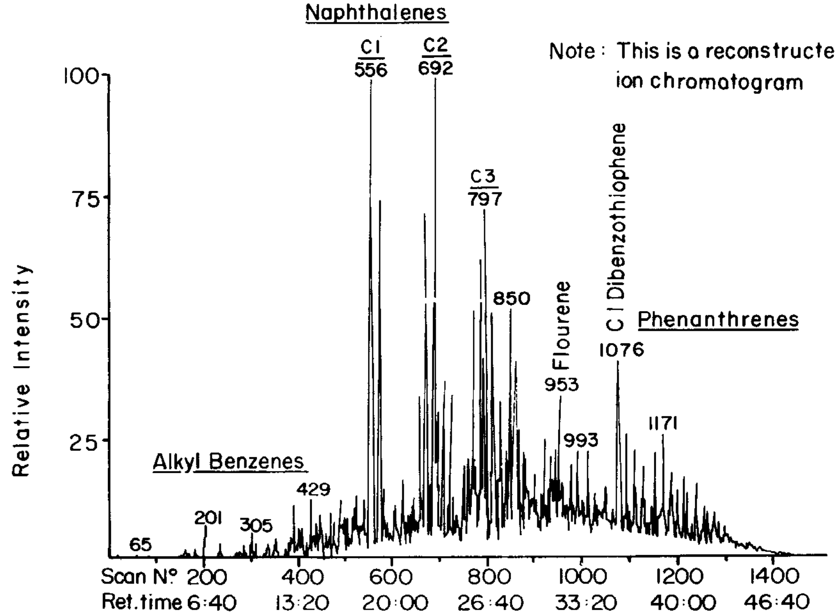


FIG. 1. Apparatus for separation of aliphatics from aromatics.

of the eluant under atmospheric pressure yielded nearly colorless saturated paraffinic hydrocarbons (76 mL). Development was continued with ethanol, and the eluate was collected in 10 fractions of 50 cm^3 each (ASTM standard D936-55 and D2003-64). Distillation of the eluant yielded a pale yellow liquid containing aromatic compounds (24 mL).

The aromatic fraction (24 mL) was subjected to further separation by liquid chromatography by using a double jacketed glass column (200 cm length, 1 cm i.d.) with a fritted disc at the bottom. The column was packed with 50 g activated silica gel (400 mesh of 30 μm diameter, product of E. Merck, Darmstadt, Germany). The gel was activated according to the previous mentioned procedure. Several fractions were collected after using different eluants of 200 cm^3 each successively , e.g., *n*-hexane, 10% volume benzene in hexane; 20% volume benzene in hexane, and mixed eluant of 20% volume benzene + 20% volume diethyl ether + 60% volume ethanol. The use of silica gel for this purpose is well known (ASTM standards D936-55, D1319-61/T, D2002-64, and D2003-64).

The collected fractions of aromatics were recombined and washed with potassium hydroxide solution followed by water. The yield was 95%, i.e., 22.8 mL neutral + basic aromatics. The gas chromatogram of this material is shown in Fig. 2.



Note : This is a reconstructed ion chromatogram

FIG. 2. Chromatogram of the neutral + basic aromatic fraction.

TABLE 2
Results of Gas Chromatography–Mass Spectrometric Analysis of the Neutral + Basic Aromatic Fraction

Class of compound	Digital counts	Area % of aromatics
1. Monoaromatics:		
C_3–C_6 Alkyl benzenes	36,735	3.68
C_1–C_5 Benzothiophenes	142,132	14.24
2. Biaromatics:		
C_0–C_4 Naphthalenes	527,197	52.81
C_0–C_4 Dibenzothiophenes	143,535	14.38
C_0–C_4 Fluorenes	41,103	4.12
C_1–Biphenyls + dibenzofuran	40,620	4.07
3. Triaromatics:		
C_0–C_4 Phenanthrenes	61,908	6.20
C_0–C_4 Pyrenes/fluoranthrenes	5,124	0.51
Total		100.00

Gas Chromatography–Mass Spectrometric Analysis

GC/MS analyses of the neutral + basic aromatics were kindly provided by Globel Geochemistry Corporation, 6169 Eaton Ave., Canoga Park, California 91303-2194, USA. Results are given in Table 2. The analyses illustrate the types (mono-, bi-, and tricyclic aromatics) and the area percentage of aromatic groups.

High Performance Liquid Chromatographic Analysis

The neutral + basic aromatic fraction was separated by preparative scale high performance liquid chromatography (Shimadzu LC-4A, HPLC) by using a preparative stainless steel column (25 cm length, 2.12 cm i.d.) containing the normal phase SIL Shipmac. The injected sample was dissolved in tetrahydrofuran and eluted with pentane. Collected fractions were distilled to remove the solvent. The aromatic fractions from this operation were identified qualitatively by GC/MS. The results given in Table 3

TABLE 3
Qualitative Analysis of the Neutral + Basic Aromatic Compounds by High Performance Liquid Chromatography (HPLC)

Type of nuclear aromatic	Molecular weight	Number of components
Mono	120–204	49
Bi	128–240	50
Tri	159–226	5

indicate the type of nuclear aromatic according to the number of rings (mono-, bi-, and tricyclic aromatics); the amount and each type present were not determined.

Each type of nuclear aromatic fraction was separated into smaller fractions by HPLC on a preparative ODS-C_{18} reverse phase column (Zorbax, 25 cm length, 2.12 cm, i.e., 880952-102 volume) as eluant. The collected fractions (500 cm^3 each) were recycled several times. Multiple extractions were performed on each of the collected fractions with methylene chloride, followed by gentle evaporation at ambient temperature and pressure to remove most of the solvent and obtain a highly concentrated solution. No internal standard was used; therefore, the data obtained are relative, not absolute concentrations.

The type and relative amount of the individual components in the separated fractions were identified and determined by analytical gas liquid chromatography supplemented by mass spectrometry (Hewlett-Packard 5993 system; for conditions, see the Experimental Section). Results of GC/MS analysis of the nonhydrocarbon aromatics are given in Table 4.

RESULTS AND DISCUSSION

Gas oil is a complex mixture of paraffinic, naphtenic, and aromatic compounds. The scheme in Fig. 3 shows the procedure applied for separating the complex mixture into various families and for separating the individual aromatic hydrocarbons and nonhydrocarbons.

The aromatic/alphatic separation used was a modification of the published separation method (ASTM D-2549, 1980, part 24, p. 464). The modifications are listed in Table 5.

The results of the analysis were 76% volume paraffinic and 24% volume aromatic compounds. These values are in good agreement with those obtained spectroscopically (from IR and NMR spectra). The aromatic fractions were subjected to further chromatographic separation on silica gel, with the different eluants listed in the Experimental Section, in order to eliminate any traces of aliphatic hydrocarbons.

With the aid of a Finnegan type 9612-4000 GC/MS apparatus (for conditions, see the Experimental Section), the mixture of neutral + basic aromatic compounds was qualitatively identified and revealed the presence of more than 112 peaks. Table 2 shows that the neutral + basic aromatic compounds consist mainly of 3.68% monocyclic aromatic hydrocarbons (C_3–C_6 alkyl benzenes), 52.81% bicyclic aromatic hydrocarbons (C_0–C_4 alkyl naphthalenes), 6.20% tricyclic aromatic hydrocarbons (C_0–C_4 alkyl penanthrenes), and 37.32% nonhydrocarbon aromatic compounds.

The components in the major HPLC peaks corresponding to aromatics were further separated into small groups (3–4 components in each) by

TABLE 4
Nonhydrocarbon Aromatic Compounds Found in the Gas Oil

No.	% Area	Base peak	Mol. wt.	Formula	Name
1	0.0454	145.1	195	$C_{11}H_{13}N$	Benzonitrile, 2,6-diethyl
2	0.0652	163.2	163	$C_7H_9N_5$	*s*-Triazolo[4,3-α]pyrazine, 3-amino-5,8-dimethyl
3	0.1470	161.2	176	$C_{11}H_{12}S$	Benzo[*b*]thiophene, 7-ethyl-2-methyl
4	0.1036	175.2	190	$C_{12}H_{14}S$	Benzo[*b*]thiophene, 2,7-diethyl
5	0.1174	175.2	190	$C_{12}H_{14}S$	Benzo[*b*]thiophene, 2,3-diethyl
6	0.1133	161.2	161	C_9H_7NS	2(1*H*)-Quinolinethione
7	0.0987	155.2	171	$C_{12}H_{13}N$	Quinoline, 2-(1-methylethyl)
8	0.1388	57.2	191	$C_{12}H_{17}NO$	Morpholine, 1,2,3,4,4a,5,6,8a-Octahydro-7-methyl-4-methylene 1-(1-methyl)-,1·α,4a·α,8a·α
9	0.1581	184.2	184	$C_{13}H_{12}O$	1,1-Biphenyl, 2-methoxy
10	0.0871	198.2	198	$C_{13}H_{12}O$	Dibenzothiophene, 3-methyl
11	0.0896	198.2	198	$C_{13}H_{10}S$	Dibenzothiophene, 4-methyl
12	0.1031	198.2	198	$C_{13}H_{10}S$	Dibenzothiophene, 5-methyl
13	0.0860	198.2	198	$C_{13}H_{10}S$	Dibenzothiophene, 6-methyl
14	0.0883	197.2	212	$C_{13}H_{10}N_2O$	Benzenamine, 4-methoxy-*N*-(3-pyridinyl methylene)
15	0.1065	212.2	212	$C_{13}H_{10}N_2O$	Benzenamine, 3-methoxy-*N*-(4-pyridinyl methylene
16	0.1270	212.2	212	$C_{14}H_{12}S$	Naphtho[2,3–6]thiophene, 4,9-dimethyl
17	0.1051	212.2	212	$C_{14}H_{12}S$	Naphtho[2,3–6]thiophene, 2,7-dimethyl
18	0.0927	212.2	212	$C_{14}H_{12}S$	Naphtho[2,3–6]thiophene, 3,8-dimethyl
19	0.0867	57.2	206	$C_{15}H_{26}$	1*H*-3a,7-Methnoazulene, Octahydro-1,4,9,9-tetramethyl
20	0.0681	211.3	226	$C_{16}H_{18}O$	[1,1′-Biphenyl]-2-o1m3- (1,1-dimethylethyl)
21	0.0672	211.2	226	$C_{16}H_{18}O$	1,1′-Biphenyl,3-(1,1-dimenthylethoxy)
22	0.0700	211.2	226	$C_{16}H_{18}O$	1,1′-Biphenyl, 8-(1,1-dimenthylethoxy)
23	0.1036	166.2	194	$C_{13}H_{10}N_2$	Benzo[*c*]cinnoline, 4-methyl
24	0.0801	179.2	210	$C_{13}H_{10}N_2O$	Phenanzine, 1-methoxy
25	0.0828	226.2	226	$C_{14}H_{10}O_3$	9*H*-Xanthen-9-one, 4-methoxy
26	0.0793	226.2	226	$C_{14}H_{10}O_3$	9*H*-Xanthen-9-one, 3-methoxy
27	0.0784	226.2	226	$C_{14}H_{10}O_3$	9*H*-Xanthen-9-one, 2-methoxy
28	0.0776	226.2	226	$C_{14}H_{10}O_3$	9*H*-Xanthen-9-one, 1-methoxy

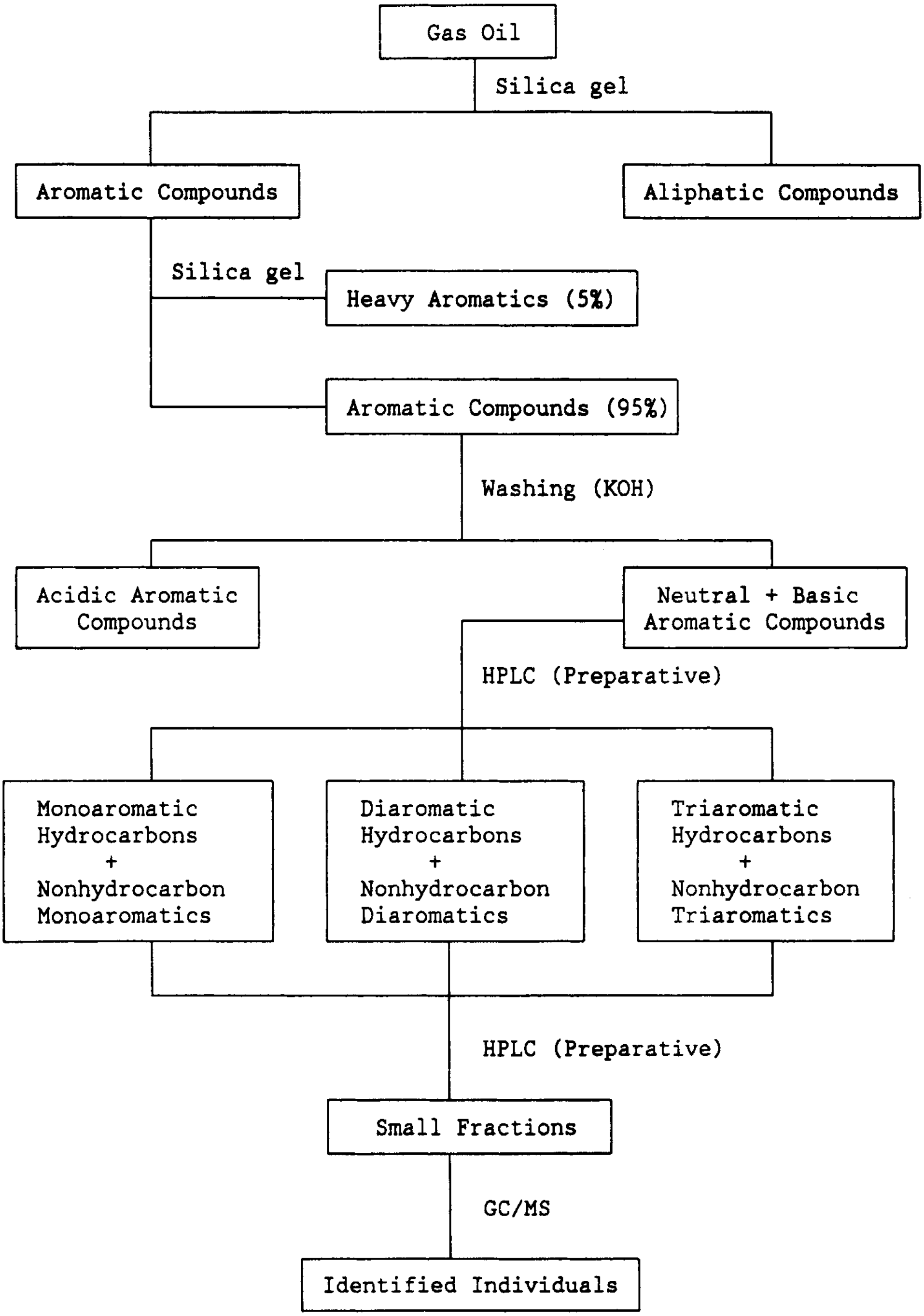


FIG. 3. Schematic diagram for separation and identification of nonhydrocarbon aromatic compounds in gas oil.

TABLE 5
ASTM D-2549 and Its Modifications

ASTM method	Modified method
Silica gel 100–200 mesh with bauxite 20-60 mesh	Silica gel 100–140 mesh (alone)
Pentane, chloroform, diethyl ether, and ethanol are used successively	*n*-Pentane as eluant for saturates and methanol for aromatics
Length of column, 114 cm	Length of column, 200 cm
Weight of sample, 10 g	Weight of sample, 100 g
Nitrogen gas is used to pressurize the column at room temperature.	Separation under atmospheric pressure at a controlled temperature (15°C)

HPLC by using a ODS-reverse phase-C_{18} column. To separate a single component from the mixture is a difficult problem, and the following parameters have to be considered: relative retention strengths (capacity ratios), resolution as a function of structure effects, and column efficiency. For the preparative system, additional factors must also be considered, such as loading capacities, reequilibration time (if a gradient is necessary), and column durability.

The individual nonhydrocarbon aromatic compounds in the separated fractions were identified by GC/MS (Hewlett-Packard 5993 system), IR, and NMR. The results are listed in Table 4. A detailed discussion of all the spectral data (available upon request from the first author) is beyond the scope of the present work.

The nonhydrocarbon aromatic series (28 compounds) accounted for 32.69% volume of this fraction of the gas oil. In this group the compounds investigated were three benzothiophenes, one benzonitrile, one pyrazine, two quinolines, four dibenzothiophenes, three naphthothiophenes, one morpholine, four biphenyloxy compounds, two benzenamines, one methnoazuline, one benzocinnoline, one phenazine, and four xanthenes.

A MS library search was made for the components and confirmed by comparison with spectra obtained for available standard samples.

The library search for 3-methyldibenzothiophene has been chosen as a representative for the following interpretations. The mass fragmentation pattern obtained for this compound is given in Table 6. The result of this search is reported in Table 7. The mass spectrum obtained for the compound and the library spectrum are given in Fig. 4. Three peaks have a similar molecular weight. The peak in the chromatogram at 48.62 min was assigned as 3-methyldibenzothiophene.

TABLE 6
The Relative Abundance of the Fragment Ions in the Mass Spectrum of 3-Methyldibenzothiophene

m/z	Relative abundance	*m/z*	Relative abundance	*m/z*	Relative abundance	*m/z*	Relative abundance
50	2.7	96	3.4	143	7.0	187	2.5
51	5.8	97	10.3	144	2.5	188	1.6
52	1.6	98	8.5	145	5.6	189	12.8
53	4.3	99	11.9	147	5.8	190	3.1
54	1.8	100	2.2	148	2.0	191	4.0
55	13.7	101	13.9	149	4.0	192	2.5
56	4.7	102	4.0	150	3.8	193	6.1
57	10.6	103	3.8	151	9.0	194	5.2
59	3.4	104	4.3	152	21.6	195	17.1
60	1.6	105	8.5	153	13.7	196	13.0
61	2.5	106	2.7	154	3.4	196	65.8
62	2.7	107	2.9	155	5.6	198	100.0
63	7.4	109	4.9	156	3.6	199	19.6
64	1.8	110	2.7	157	5.4	200	6.3
65	4.0	111	4.9	158	2.9	201	2.7
66	1.6	112	2.0	159	3.4	203	6.1
67	7.4	113	4.7	160	2.5	204	1.8
68	1.6	114	2.5	161	5.6	205	1.8
69	12.8	115	16.0	162	3.1	207	1.6
70	2.9	116	2.7	163	6.5	208	2.9
71	7.2	117	4.7	164	4.9	209	7.0
73	2.2	118	4.3	165	31.7	210	10.3
74	4.0	119	8.1	166	8.8	211	3.1
75	4.5	120	3.4	167	5.6	212	3.6
76	7.2	121	3.6	168	2.9	213	2.0
77	8.3	123	3.6	169	7.0	216	1.6
78	3.4	125	3.6	170	3.4	217	2.7
79	4.9	126	4.5	171	11.9	218	3.6
80	1.8	127	10.1	173	2.5	219	1.3
81	7.6	128	10.1	172	4.0	223	1.6
82	5.2	129	10.8	174	2.9	224	2.7
83	5.6	130	3.1	175	7.9	225	1.6
84	2.9	131	7.2	176	4.5	227	1.3
85	7.6	132	2.7	177	4.0	232	2.7
86	5.2	133	5.4	178	8.8	234	1.1
87	5.6	134	4.3	179	10.1	237	1.1
88	2.7	135	3.8	180	3.6	241	.9
89	6.1	137	3.4	181	16.6	242	1.3
91	9.2	138	2.2	182	4.0	246	1.1
92	2.2	139	7.9	183	9.4	252	1.1
93	5.4	140	2.2	184	3.6	262	.7
94	2.5	141	8.1	185	4.5		
95	7.2	142	4.3	186	1.6		

TABLE 7
Library Search for 3-Methyldibenzothiophene Present in Gas Oil

Nonhydrocarbon aromatic	Contamination, %	Match factor, %	Qualitative Index	Matching peaks
Dibenzothiophene 3-methyl	22.7	0.7233	641	15.2
Benzene, 1,1′-Cl-fluoro-1–2 (ethendiyl)bis,	31.7	0.8607	546	17.3
9*H*-Thioxanthene	35.9	0.7410	401	15

From Fig. 4 and Table 7 the following interpretations help greatly in determining the structure of the compound under study: (1) the relative abundance of the molecular ion for the compound is 85.81% compared with 53% of 3-methyldibenzothiophene, and 9*H*-thioxanthene (100%), taking in consideration that the fluoro compound does not exist in the neutral aromatic fraction; (2) the contamination value (22.7) is lower than that (35.9) of 9*H*-thioxanthene, which means that 3-methyldibenzothiophene is the most probable matching compound; (3) the matching peak of 3-methyldibenzothiophene (15.2%) is higher than that (15%) of 9*H*-thioxanthene. From the above comparison, it is clear that 3-methylidbenzothiophene is the most likely match for the sample.

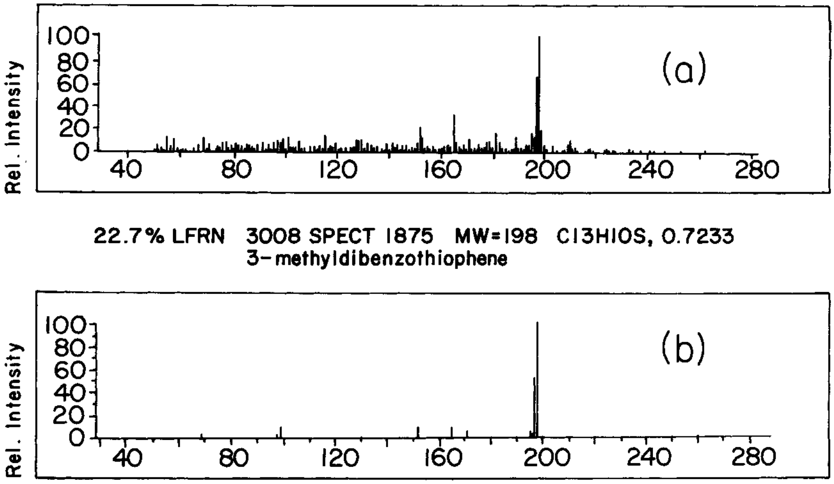


FIG. 4. Spectrum of 3-methyldibenzothiophene (a) and library spectrum (b).

The same procedure was applied for identifying other nonhydrocarbon aromatics.

In addition to yielding basic information concerning the composition of petroleum, this work illustrates the application of modern physical separation processes and spectroscopic methods of identification to the analysis of a very complex mixture of paraffinic and aromatic compounds.

Refiners dealing with gas oil will find the data in this work helpful in quick evaluation of feedstock and product of Kuwait petroleum as well as in improving processing operations through a better capacity to follow the chemical reactions involved.

Further quantitative developments on neutral + basic aromatic compounds are in progress in our laboratory, using the HPLC procedure described here. This work will be extended to obtain compositional data which could be compared for several samples of gas oil from different sources.

Acknowledgment

The authors wish to express their gratitude to Kuwait Foundation for the Advancement of Sciences for financial support.

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Received by editor June 29, 1990
Revised June 4, 1991