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## DETERMINATION OF SOME POLYNUCLEAR AROMATIC HYDROCARBONS IN NEW, USED AND REGENERATED OILS

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### SUMMARY

The levels of polynuclear aromatic hydrocarbons (PAHs) in oils were determined using a three-step process: extraction and concentration by charge-transfer liquid chromatography on an improved tetrachlorophthalimidopropyl-bonded silica; oxidation of the thiophenes, which are extracted and concentrated at the same time, and separation of the resulting sulphones from the PAHs by adsorption chromatography on bare silica; and separation of the PAH fraction of improved purity on tetrachlorophthalimidopropyl-bonded silica. The recoveries for six PAHs were greater than 70%, except for indeno[1,2,3-*cd*]pyrene (28%), and they can be determined in new and used lubricating and regenerated oils.

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### INTRODUCTION

Charge-transfer liquid chromatography is mainly used for the separation of polynuclear aromatic hydrocarbons (PAHs)<sup>1–3</sup> and has been the subject of a number of reviews<sup>4–6</sup>. The retention of a solute in charge-transfer liquid chromatography is based on  $\pi$ - $\pi$  interactions between the solutes and the stationary phase<sup>7–9</sup> and the main parameter governing PAH retention is the density of their electronic cloud constituted by delocalized  $\pi$ -electrons. Recently, we defined a series of structural parameters which leads to a rapid determination of the retention order of unsubstituted and substituted PAHs<sup>10</sup>.

One of the interesting aspects of charge-transfer liquid chromatography is the high capacity factors for PAHs achieved using a  $\pi$ -electron acceptor-bonded silica with an apolar mobile phase. We improved the synthesis of tetrachlorophthalimidopropyl-bonded silica<sup>11</sup>, a support described by Holstein in 1981<sup>12</sup>, and on this improved

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support, with isooctane as mobile phase, the capacity factors were higher than 75 for four-ring PAHs, 700 for five-ring PAHs and 2000 for six-ring PAHs<sup>11</sup>.

An obvious application of charge-transfer liquid chromatography on tetrachlorophthalimidopropyl-bonded silica is the extraction and concentration of PAHs in apolar media such as oils. These are mainly composed of saturated hydrocarbons which have no delocalized  $\pi$ -electrons, and are not retained by charge-transfer liquid chromatography (olefinic hydrocarbons are very weakly retained).

In comparison with the usual methods of determination, this approach partially resolved the following problems: the low recovery obtained with liquid-liquid extraction<sup>13-15</sup> or with adsorption chromatography<sup>1,16</sup>; the low solubility of the samples in aqueous mobile phases used for reversed-phase liquid chromatography<sup>17,18</sup>; the low volatility of real samples when they are analysed by gas chromatography; and the fact that fluorescence detection is much more selective than flame ionization detection<sup>19,20</sup> and more sensitive than mass spectrometry<sup>21</sup>.

However, the determination of the concentration of PAHs in oils on tetrachlorophthalimidopropyl-bonded silica requires an intermediate step, *viz.*, the separation of PAHs from polynuclear derivatives with a heterocycle, especially thiophenes, which are also strongly retained by charge-transfer liquid chromatography.

This approach has been applied to the measurement of the concentrations of PAHs in a series of new, used and regenerated lubricating oils and the results are reported in this paper.

## EXPERIMENTAL

### *Apparatus*

A Model 5020 liquid chromatograph (Varian, Palo Alto, CA, U.S.A.) was used in conjunction with an LDC Spectromonitor III detector (Milton Roy, LDC Division, Riviera Beach, FL, U.S.A.) or with an LS4 spectrofluorometric detector (Perkin-Elmer, Norwalk, CT, U.S.A.). The integrator was a Chromatopac C-R3A (Shimadzu, Kyoto, Japan). The temperature was set at  $40 \pm 0.1^\circ\text{C}$  with a constant-temperature water-bath.

### *Stationary phases*

Two chromatographic columns were packed with tetrachlorophthalimidopropyl-bonded silica, which was prepared as described previously<sup>11</sup>. The ligand density was  $2.7 \mu\text{mol m}^{-2}$ . The dimensions of the column used for the concentration step were  $15 \text{ cm} \times 6.9 \text{ mm I.D.}$  The dimensions of the analytical column were  $25 \text{ cm} \times 4.6 \text{ mm I.D.}$

The columns were polished stainless-steel tubes fitted with a  $2\text{-}\mu\text{m}$  porous stainless-steel frit. They were packed by using the classical slurry packing technique: the bonded silica was suspended in carbon tetrachloride-methyl iodide (67:33, v/v) and the slurry was pushed with ethanol into the chromatographic column for 30 min under a 500-bar pressure.

C<sub>18</sub> columns ( $5 \text{ cm} \times 4.6 \text{ mm I.D.}$ ; particle size  $3 \mu\text{m}$ ) were provided by Chrompack (Middelburg, The Netherlands).

### Chemicals

The porous silica used to synthesize the bonded electron acceptor phase was 10- $\mu\text{m}$  Lichrosorb Si 100 (pore diameter 100 Å; specific surface area 350  $\text{m}^2 \text{g}^{-1}$ ) (Merck, Darmstadt, F.R.G.). Adsorption chromatography was carried out on 63–200- $\mu\text{m}$  silica gel 100 (Merck).

PAHs were provided by the Community Bureau of Reference (Brussels, Belgium), Fluka (Buchs, Switzerland) and Janssen Chimica (Beerse, Belgium), the thiophenes were from Aldrich (Milwaukee, WI, U.S.A.) and the oils and the additives were obtained from Régie Renault (Rueil-Malmaison, France).

Isooctane and methylene chloride were purchased from Rathburn (Walkerburn, U.K.), toluene, acetonitrile and hydrogen peroxide from Merck and acetic acid from Prolabo (Paris, France). Water was doubly distilled.

## RESULTS

### Procedure

The determination of PAHs in apolar media was accomplished in three steps, as follows.

**Extraction and concentration.** The column was initially equilibrated with isooctane. Then, an apolar matrix,  $V = 300 \text{ ml}$ , was percolated into the column, where  $V = V_1 + V_2$ ,  $V_1$  being the volume of oil and  $V_2$  the volume of isooctane added. The addition of isooctane was necessary to reduce the viscosity of the percolated medium, *i.e.*, to reduce the pressure drop. The strongly fixed PAHs and eventually other molecules (*e.g.*, thiophenes, see later) were eluted from the column by back-flushing with methylene chloride. This solvent has two interesting characteristics: (1) from the fixation isotherms, its affinity with respect to the tetrachlorophthalimidopropyl groups, and consequently its eluting power, are very high<sup>10</sup>, so the PAHs are eluted very rapidly; and (2) it is very volatile and the concentration of the PAH-containing fraction (to  $V_3$ ) is rapid. In this work, the column was completely eluted with 50 ml of methylene chloride, the eluate being evaporated to 0.5 ml: the enrichment factor was equal to  $V_1/V_3 = 300$ .

**Separation of PAHs and thiophenes.** In the previous step, the PAHs were

TABLE I  
RETENTION OF PAHs AND THIOPHENES ON TETRACHLOROPHTHALIMIDOPROPYL-BONDED SILICA

Column, 25 cm  $\times$  4.6 mm I.D.; stationary phase, tetrachlorophthalimidopropyl-bonded LiChrosorb Si 100 (10  $\mu\text{m}$ ), ligand density 2.7  $\mu\text{mol m}^{-2}$ ; mobile phase, isooctane–methylene chloride; flow-rate, 1.2  $\text{ml min}^{-1}$ ; detection, UV absorbance at 254 nm. Solutes: Fl = fluorene; DBT = dibenzothiophene; Ant = anthracene; Ft = fluoranthene; BNT = benzo[*b*]naphtho[2,1-*d*]thiophene; B[*a*]A = benz[*a*]anthracene.

Isooctane–methylene chloride	Fl	DBT	Ant	Ft	BNT	B[ <i>a</i> ]A
70:30 (v/v)	—	3.0	3.2	6.5	7.4	7.6
80:20 (v/v)	—	4.9	5.3	11.2	15.0	15.7
90:10 (v/v)	—	11.0	11.6	24.6	39	41
100:0 (v/v)	20	44	50 <sup>a</sup>	80 <sup>a</sup>	—	—

<sup>a</sup> Extrapolated values.

concentrated, but also were polynuclear derivatives that contain a heterocycle, especially thiophenes (Table I). This is due to both the electron pairs on the sulphur atom and  $\pi$ -electron delocalization. In a ring containing four carbon atoms plus one sulphur atom, the  $\pi$ -electron delocalization is the same as that for a benzene ring and, for example, a four-benzene-ring PAH and a four-ring thiophene have almost the same retention on this stationary phase. Hence the separation of PAHs and thiophenes was necessary and required the oxidation of the thiophenes to sulphones (after an intermediate oxidation to sulphoxides). Sulphones are very polar whereas the PAHs are apolar, so the separation of PAHs from sulphones can be achieved by adsorption chromatography on bare silica. The oxidation procedure was as follows<sup>22</sup>: the concentrated PAH mixture was added to 100 ml of toluene-acetic acid (50:50, v/v), the solution was brought to reflux, 50 ml of hydrogen peroxide were added during 1 h and reflux was continued for 16 h. The aqueous and organic phases were separated and the toluene phase washed with 250 ml of distilled water, the resulting aqueous phase then being washed with 50 ml of toluene. The 100 ml of toluene (50 ml from the initial mixture + 50 ml from the washing of water) were evaporated to 10 ml, which were percolated on the top of a bare silica column (40 g of silica). The PAHs were eluted with 100 ml of water-saturated toluene, and the eluate was evaporated to less than 0.5 ml.

*Separation of PAHs.* This step was effected by charge-transfer liquid chromatography on tetrachlorophthalimidopropyl-bonded silica. The mobile phase was isooctane-methylene chloride with gradient elution. The temperature was maintained at  $40 \pm 0.1^\circ\text{C}$  to improve the resolution and keep the duration of analysis short. A spectrofluorimetric detector was used, which was time-programmable with respect to the excitation and emission wavelengths to improve the selectivity and the sensitivity. Of course, this detection method needed a very reproducible separation. For the determination of PAH concentration, the PAH extract was diluted to a determined volume, *e.g.*, 0.5, 1 or 2 ml.

TABLE II

ADDED AND MEASURED AMOUNTS OF FLUORANTHENE, BENZO[k]FLUORANTHENE AND BENZO[a]PYRENE AND RECOVERIES OF THE EXTRACTION STEP

Column: 15 cm  $\times$  6.9 mm I.D. Abbreviations as in Table I.

Percolated solution	PAH	Amount (mg)		Recovery (%)
		Added	Measured	
300 ml isooctane				
+ 13.2 mg l <sup>-1</sup> Ft	Ft	3.96	3.63	91.7
1.45 mg l <sup>-1</sup> B[k]F	B[k]F	0.43	0.47	> 100
5.10 mg l <sup>-1</sup> B[a]P	B[a]P	1.53	1.48	96.7
300 ml standard	Ft	3.96	3.86	97.5
solution	B[k]F	0.43	0.50	> 100
+ 50 ml Neutral 350	B[a]P	1.53	1.47	96.1
300 ml standard	Ft	3.96	3.86	97.5
solution	B[k]F	0.43	0.52	> 100
+ 50 ml Neutral 100	B[a]P	1.53	1.44	94.1

*Comments*

*Extraction and concentration.* The volume percolated was established from the dead volume of the column (4.2 ml) and the retention of the least retained PAH, fluoranthene. Its capacity factor on tetrachlorophthalimidopropyl-bonded silica, with pure isooctane as the mobile phase, is 75; consequently, if an almost 100% recovery is required, the volume percolated has to be lower than  $4.2 \times 75 = 315$  ml (the column will never be overloaded, owing to the low concentrations of PAH in oils).

We chose 300 ml, and we verified the quantitiveness of the process with three solutions: a "standard" solution composed of 4.4 mg of fluoranthene, 0.48 mg of benzo[*k*]fluoranthene and 1.7 mg of benzo[*a*]pyrene in 100 ml of pure isooctane; 300 ml of the "standard" solution + 50 ml of new Neutral 350; and 300 ml of the "standard" solution + 50 ml of new Neutral 100. Neutral 350 and Neutral 100 are base oils containing aromatic compounds and without any additives. Table II presents the results and, within the experimental error, the extraction and concentration step can be considered to be quantitative. The worst recovery was 92% and the precision was 5%.

For lubricating oils we had to determine the influence of the additives that they contain. None of them is polyaromatic and they were not strongly retained on tetrachlorophthalimidopropyl-bonded silica (Table III). The highest capacity factor was lower than 3 with isooctane as the mobile phase. Consequently, the additives were not retained during the concentration step.

All PAHs are eluted from the column with 50 ml of methylene chloride and the support is then perfectly clean. In addition, the evaporation of methylene chloride does not add any interfering peaks. Under these conditions, the tetrachlorophthalimidopropyl-bonded silica was stable for more than 1 year.

*Separation of PAHs and thiophenes.* Sulphur is the third most abundant element in mineral oils, after carbon and hydrogen, and thiophenes are always present in oils.

TABLE III

FUNCTION AND RETENTION OF OIL ADDITIVES ON TETRACHLOROPHTHALIMIDOPROPYL-BONDED SILICA WITH PURE ISOOCTANE

<i>Additive</i>	<i>Function</i>	<i>k'</i>
MDTP <sup>a</sup>	Anti-oxidant	0.75
Zn di-2-octyldithiophosphate	Anti-wear	0.87
Thiophosphate <sup>b</sup>	Detergent	<2
Carbonate <sup>b</sup>	Detergent	<2
Phenol <sup>b</sup>	Anti-oxidant	<2
Polyisobutylene <sup>b</sup>	Viscosity improver	<2
Poly(ethylene-propylene)	Viscosity improver	<3
Alkyl polymethacrylate	Viscosity improver	<3
Chlorinated alkane	High-pressure resistance	≈0
Chlorinated hydrocarbons <sup>c</sup>	High-pressure resistance	<1.4
Sulphurized hydrocarbons <sup>c</sup>	High-pressure resistance	<1.4
Fat <sup>c</sup>	High-pressure resistance	<1.4
Sulphurized ester	High-pressure resistance	1.3

<sup>a</sup> MDTP = 4,4'-methylenebis(2,6-di-*tert.*-butylphenol).

<sup>b</sup> Additive mixture; the capacity factor of the most retained is less than 2.

<sup>c</sup> Additive mixture; the capacity factor of the most retained is less than 3.

The sulphur-containing compounds can represent up to 10% of the oil matrix<sup>23</sup> and, owing to their strong  $\pi$ -electron delocalization, thiophenes are retained similarly to PAHs (Table I).

The oxidation process has been fully described previously<sup>22</sup>, and Fig. 1 shows its efficiency. For a used thermal oil, fluoranthene, benzo[*k*]fluoranthenes, benzo[*a*]pyrene and possibly benzo[*ghi*]perylene can be determined, but only after the separation of PAHs from thiophenes.

**Separation of PAHs.** The separation of PAHs was achieved by charge-transfer liquid chromatography on tetrachlorophthalimidopropyl-bonded silica with isooctane–methylene chloride as the mobile phase. An elution gradient programme from 30% to 100% methylene chloride was used. Fig. 2 shows the separation of six PAHs: fluoranthene, benzo[*k*]- and benzo[*b*]fluoranthene, benzo[*a*]pyrene, indeno[1,2,3-*cd*]pyrene and benzo[*ghi*]perylene. These PAHs are the most toxic in the National Bureau of Standards list<sup>24</sup>, and we decided to determine the concentration of only these six in order to simplify the analytical problem. The separation on tetrachlorophthalimidopropyl-bonded silica (Fig. 2a) and on C<sub>18</sub>-bonded silica (Fig. 2b) can be compared. Although the selectivity was higher in charge-transfer

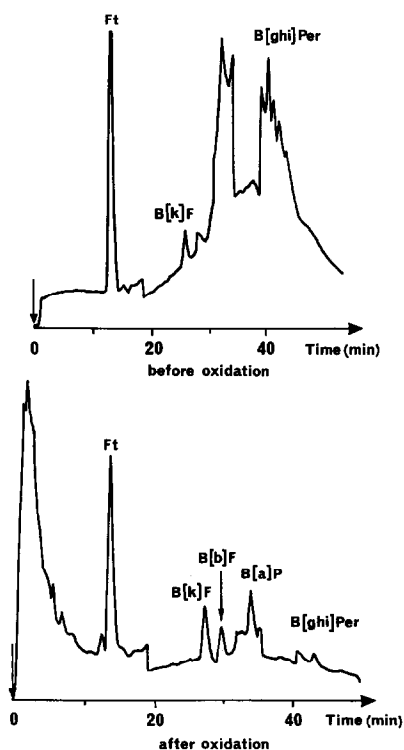


Fig. 1. Separation of PAHs in a used lubricating oil before and after oxidation of thiophenes. Chromatographic conditions as in Table I. Mobile phase: 0–13 min, isooctane–methylene chloride (70:30, v/v); 13–23 min, 70:30 to 50:50 (v/v); 23–37 min, 50:50 to 0:100 (v/v). Solutes: Ft = fluoranthene; B[*k*]F = benzo[*k*]fluoranthene; B[*b*] = benzo[*b*]fluoranthene; B[*a*]P = benzo[*a*]pyrene; B[*ghi*]Per = benzo[*ghi*]perylene; ? = unidentified.

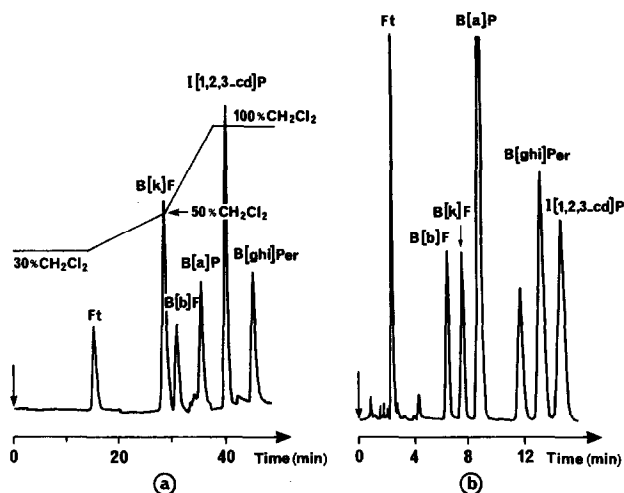


Fig. 2. Separation of six PAHs, (a) by charge-transfer liquid chromatography on tetrachlorophthalimido-propyl-bonded silica and (b) by reversed-phase liquid chromatography. (a) Chromatographic conditions as in Table I. Detection: spectrofluorimetry: 0–20 min,  $\lambda_{ex} = 278$  nm,  $\lambda_{em} = 440$  nm; 20–30 min,  $\lambda_{ex} = 308$  nm,  $\lambda_{em} = 411$  nm; 30–32 min,  $\lambda_{ex} = 302$  nm,  $\lambda_{em} = 437$  nm; 32–37 min,  $\lambda_{ex} = 285$  nm,  $\lambda_{em} = 410$  nm; 37–42 min,  $\lambda_{ex} = 277$  nm,  $\lambda_{em} = 504$  nm; 42–50 min,  $\lambda_{ex} = 273$  nm,  $\lambda_{em} = 412$  nm. (b) Column, 5 cm  $\times$  4.6 mm I.D.; stationary phase,  $C_{18}$ , particle diameter 3  $\mu$ m; mobile phase, acetonitrile–water (75:25, v/v); flow-rate, 1 ml min<sup>-1</sup>; detection, UV absorbance at 254 nm. Solutes as in Fig. 1; I[1,2,3-cd]P = indeno[1,2,3-cd]pyrene.

liquid chromatography, the resolutions were comparable and the duration of analysis was shorter using reversed-phase liquid chromatography. In addition, the separation was obtained with isocratic elution, so no column regeneration was needed. However, the solubility of the PAHs is very low in an aqueous medium, and is even lower for real samples. Consequently, charge-transfer liquid chromatography was used for the efficient and reproducible separation of PAH.

Detection was effected with a spectrofluorimetric detector that was time-programmable with respect to the excitation and emission wavelengths in order to improve both the selectivity and sensitivity simultaneously. Fig. 2a shows the wavelength programme and Table IV gives the detection limits. PAHs at concentrations lower than 1  $\mu$ g l<sup>-1</sup> (ppb) can be identified and determined.

TABLE IV

## DETECTION LIMITS FOR SIX PAHs

Chromatographic conditions as in Fig. 2. Abbreviations as in Table I.

PAH	Detection limit ( $\mu$ g l <sup>-1</sup> )
Ft	10
B[k]F	0.26
B[b]F	15
B[a]P	1.5
I[1,2,3-cd]P	14
B[ghi]Per	29

TABLE V

RECOVERIES FOR SIX PAHs FOR THE FULL DETERMINATION PROCEDURE (WITH A 95% CONFIDENCE INTERVAL)

Abbreviations as in Table I.

PAH	Recovery (%)
Ft	72 ± 4
B[k]F	96 ± 8
B[b]F	97 ± 5
B[a]P	71 ± 28
I[1,2,3-cd]P	28 ± 19
B[ghi]Per	82 ± 2

### Recoveries

The recoveries were determined by adding known amounts of the target PAHs to a new lubricating oil (this oil was new in order to reduce the total amount of PAHs, *i.e.*, to be close to the detection limits). For each PAH, a linear relationship between the added and measured amounts was observed, the recovery being equal to the slope.

Table V gives the recoveries determined with a 95% confidence interval. The results obtained for benzofluoranthene are excellent and for fluoranthene, benzo[*a*]pyrene and benzo[*ghi*]perylene they are satisfactory. However, for indeno[1,2,3-*cd*]pyrene the recovery is very low. An explanation could be degradation of this PAH during the oxidation of thiopenes. For both indeno[1,2,3-*cd*]pyrene and benzo[*a*]pyrene the calculated recovery is imprecise owing to an unstable baseline during their detection.

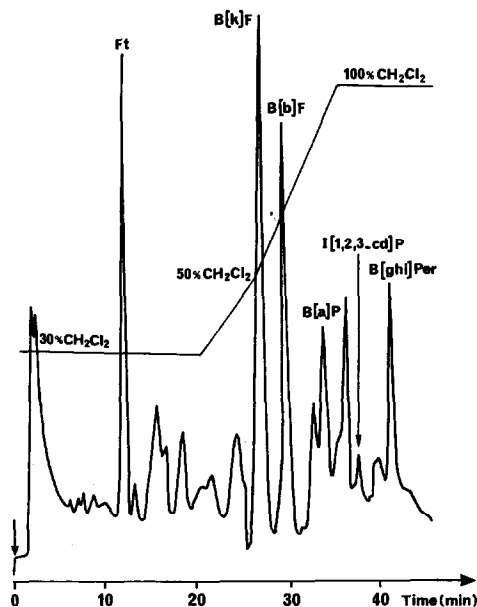


Fig. 3. Determination of PAHs in regenerated oil. Chromatographic conditions as in Fig. 2.



TABLE VI  
MEASURED PAH CONCENTRATIONS IN A NEW LUBRICATING OIL, IN TWO USED LUBRICATING OILS AND IN A REGENERATED OIL (WITH A 95% CONFIDENCE INTERVAL)  
Abbreviations as in Table I.

Oil	Concentration (ppb)					
	<i>Ft</i>	<i>B[k]F</i>	<i>B[b]F</i>	<i>B[a]P</i>	<i>I[1,2,3-cd]P</i>	<i>B[ghi]Per</i>
New lubricating oil	0.90 ±0.003	0.063 ±0.0011	0.017 ±0.003	0.011 ±0.003	0.0264 ±0.0119	3.8 ±3.6
Used lubricating oil A	336 ±58	0.24 ±0.015	43 ±4	8.64 ±1.69	16.0 ±5.0	7.91 ±1.02
Used lubricating oil B	164 ±23	0.097 ±0.017	23 ±3	2.51 ±0.6	18.6 ±5.6	4.64 ±0.26
Regenerated oil	108 ±27	5.15 ±0.22	84.0 ±8.7	14.55 ±2.25	44.4 ±21.7	83.94 ±12.67

### Sensitivity

With a signal-to-noise ratio of 3, the sensitivity ranged from 50 ppt to 5 ppb, *i.e.*, fluoranthene  $1 \text{ ng l}^{-1}$ , benzo[*k*]fluoranthene  $0.05 \text{ ng l}^{-1}$ , benzo[*b*]fluoranthene  $2 \text{ ng l}^{-1}$ , benzo[*a*]pyrene  $0.3 \text{ ng l}^{-1}$ , indeno[1,2,3-*cd*]pyrene  $5 \text{ ng l}^{-1}$  and benzo[*ghi*]perylene  $5 \text{ ng l}^{-1}$ .

### PAH concentrations in oils

PAHs were determined in four oils: a new lubricating oil, used lubricating oils A and B and a regenerated oil. For economic reasons, oil regeneration is becoming increasingly common, but leads to an increase in PAH concentrations. Hence the determination of PAHs in regenerated oils is essential.

Fig. 3 shows the separation of the PAHs detected in the regenerated oil and Table VI gives the measured PAH concentrations. The peaks were mainly identified by comparison with the retention times of standards.

Although in the new oil the PAH concentrations were very low and determined only inaccurately, they were much higher in the used oils and could be measured with good accuracy. Several concentrations are particularly high and justify the interest in this study. The PAH concentrations in regenerated oils are particularly disturbing, and, from Fig. 3, various PAHs could be identified and determined.

### CONCLUSION

Charge-transfer liquid chromatography appears to be a satisfactory method for the determination of PAHs in oils: the extraction is selective and quantitative (very high capacity factors) owing to the mechanism based on the  $\pi$ - $\pi$  interactions between the PAHs and the tetrachlorophthalimidopropyl-bonded silica; and the high selectivity and adequate efficiency allow chromatographic separation on the same

chromatographic support, *e.g.*, with an organic mobile phase (isooctane–methylene chloride) in which the PAH extract is soluble.

The problem of the simultaneous extraction and concentration of thiophenes was solved by their oxidation to polar sulphones, which were separated from the apolar PAHs by adsorption chromatography.

PAH concentrations in lubricating and regenerated oils were determined and the results show that these oils are unsafe.

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