The Synthesis of Polycyclic Thiophenes Derived From Phenanthrene Intermediates

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The synthesis of benzo[*b*]triphenyleno[2,1-*d*]thiophene (**9**), benzo[*b*]triphenyleno[1,2-*d*]thiophene (**13**), 5-methylbenz[7,8]anthra[2,1-*b*]thiophene (**17**), 1-methylchryseno[3,4-*b*]thiophene (**18**), triphenyleno[1,2-*c*]dibenzothiophene (**22**), triphenyleno[2,1-*a*]dibenzothiophene (**26**), triphenyleno[1,2-*a*]dibenzothiophene (**29**), and triphenyleno[2,1-*b*]dibenzothiophene (**30**) is described.

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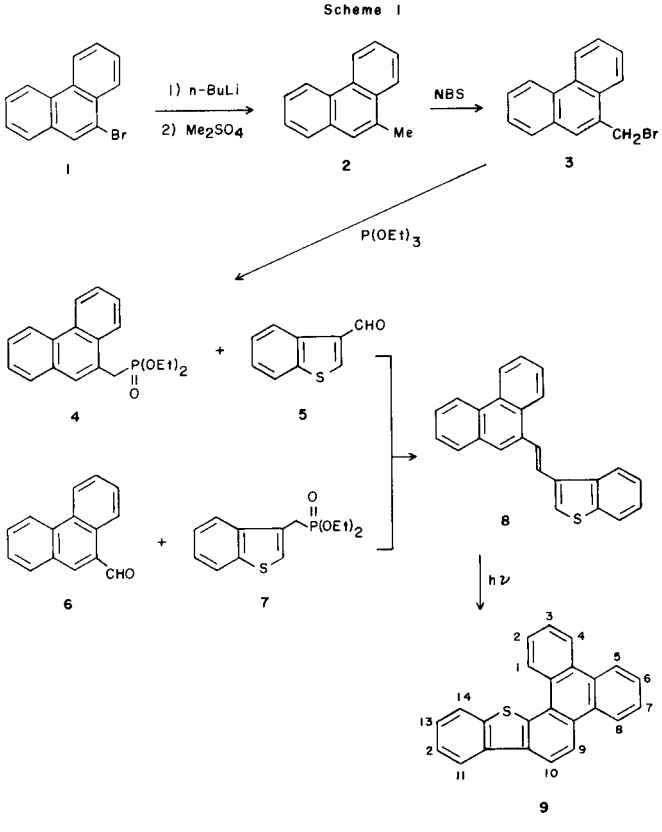
In this paper we report the synthesis of some penta-, hexa- and heptacyclic thiophenes prepared by cyclizing the phenanthrene ring system with benzo[*b*]thiophene and dibenzothiophene as a part of our continuing study [2-22] directed toward providing all of the possible polycyclic thiophenes suspected of occurring in coal liquids or related coal derived products. A number of polycyclic thiophenes have already been shown to be present in coal-derived products [23].

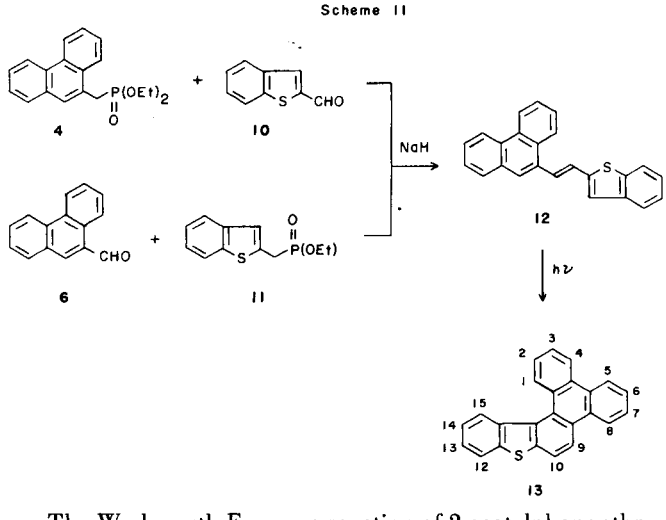
We have observed that the photocyclization reaction of aromatic vinyl substituted benzo[*b*]thiophenes is a very useful method of forming polycyclic thiophenes. Thus, benzo[*b*]triphenyleno[2,1-*d*]thiophene (**9**) was prepared by photocyclizing 1-(3′-benzo[*b*]thienyl)-2-(9″-phenanthryl)-ethene (**8**) in 73% yield. The synthesis of **8** can be accomplished by the Wadsworth-Emmons reaction between either diethyl 9-phenanthrylmethylphosphonate (**4**) and benzo[*b*]thiophene-3-carboxaldehyde (**5**) or diethyl 3-benzo[*b*]thenylphosphonate (**7**) [3] and phenanthrene-9-carboxaldehyde (**6**) in 69% and 45% yields, respectively (Scheme I).

Scheme I



In a similar manner, benzo[*b*]triphenyleno[1,2-*d*]thiophene (**13**) was synthesized by photocyclization of 1-(2′-benzo[*b*]-thienyl)-2-(9″-phenanthryl)ethene (**12**) in 50% yield. Again, **12** can be prepared by the Wadsworth-Emmons reaction between either **4** and benzo[*b*]thiophene-2-carboxaldehyde (**10**) or **6** and diethyl 2-benzo[*b*]thenylphosphonate (**11**) [12] (Scheme II).

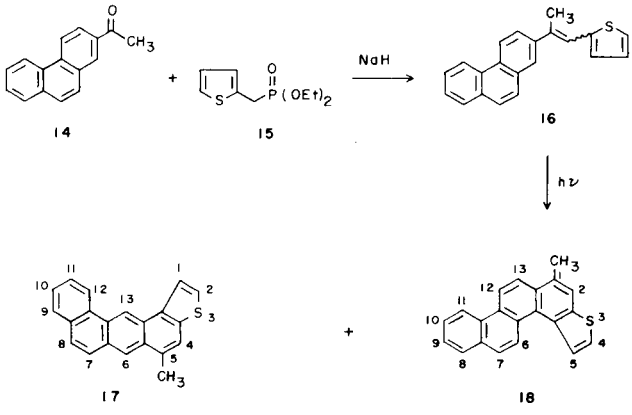
Scheme II



The Wadsworth-Emmons reaction of 2-acetylphenanthrene (**14**) and diethyl 2-thenylphosphonate (**15**) formed 1-methyl-1-(2′-phenanthryl)-2-(2″-thienyl)ethene (**16**) in 50% yield. The photocyclization of **16** produced both

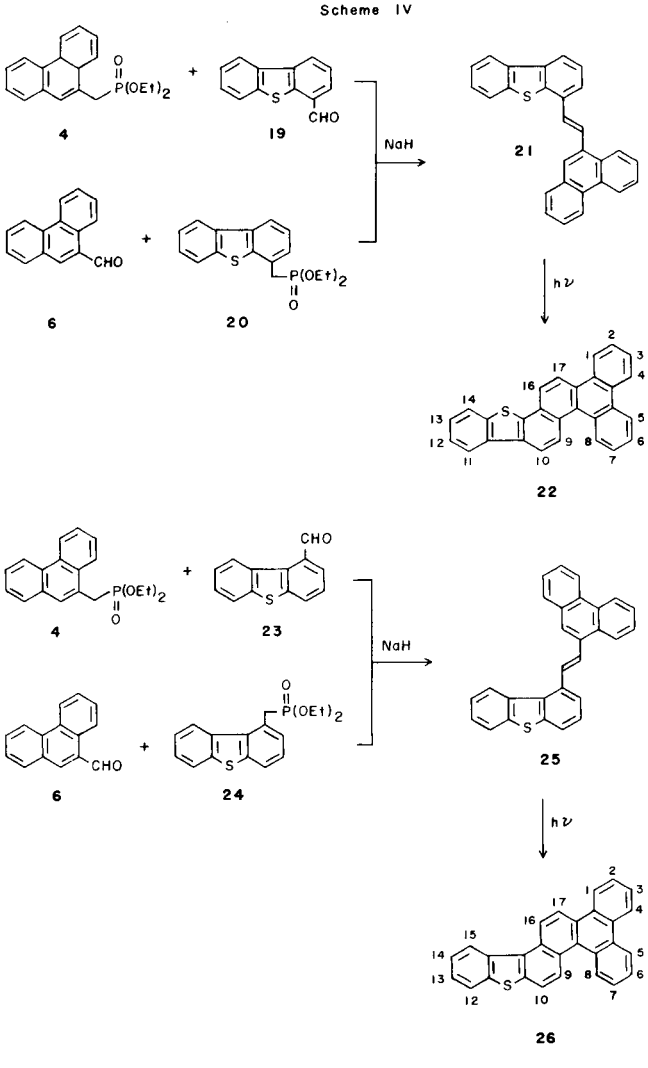
5-methylbenz[7,8]anthra[2,1-*b*]thiophene (**17**) and 1-methylchryseno[3,4-*b*]thiophene (**18**) in 31% and 34% yields, respectively (Scheme III).

Scheme III



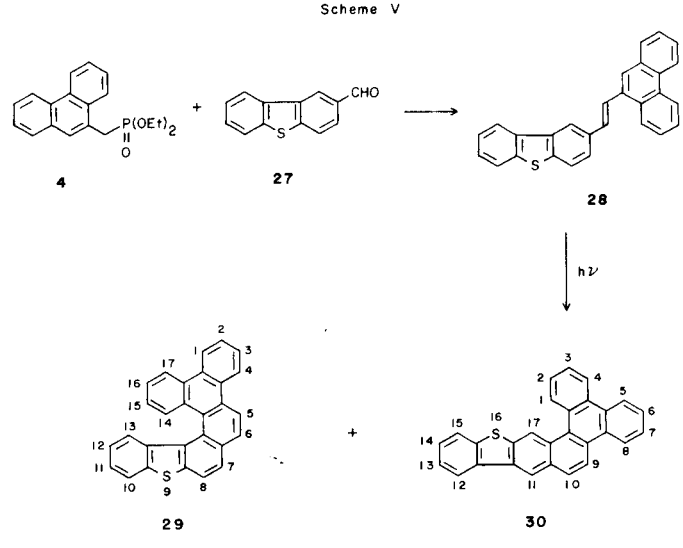
Triphenyleno[1,2-*c*]dibenzothiophene (**22**) was synthesized in 78% yield by the photocyclization of 1-(4'-dibenzothienyl)-2-(9''-phenanthryl)ethene (**21**). Compound **21** can be made by the Wadsworth-Emmons reaction of either **4** with dibenzothiophene-4-carboxaldehyde (**19**) [24] or **6** with diethyl 4-dibenzothenylphosphonate (**20**) [19] in 52% and 64% yield, respectively. Triphenyleno[2,1-*a*]dibenzothiophene (**26**) was produced by the photolysis of 1-(1'-dibenzothienyl)-2-(9''-phenanthryl)ethene (**25**). Compound **25** was synthesized *via* the Wadsworth-Emmons reaction of either **4** with dibenzothiophene-1-carboxaldehyde (**23**) [18] or **6** with diethyl 1-dibenzothenylphosphonate (**24**) [18] in 60% and 57% yields, respectively (Scheme IV).

Scheme IV



Both triphenyleno[1,2-*a*]dibenzothiophene (**29**) and triphenyleno[2,1-*b*]dibenzothiophene (**30**) were prepared by the photocyclization of 1-(2'-dibenzothienyl)-2-(9''-phenanthryl)ethene (**28**). Compound **28** was prepared from the Wadsworth-Emmons reaction between **4** and dibenzothiophene-2-carboxaldehyde (**27**) [24] (Scheme V). The polycyclic thiophenes will be screened for mutagenic activity in the Ames test using S9 liver homogenate activation against T-98 and TA-100 and the results will be reported elsewhere.

Scheme V



EXPERIMENTAL

Melting points were determined on a Thomas Hoover melting point apparatus and are uncorrected. The ^{1}H-nmr spectra were obtained on either a Varian EM360A or a JEOL FX90Q spectrometer in the solvent indicated with TMS as the internal standard. Chemical shifts are reported in δ units. The mass spectra were obtained on a Hewlett-Packard model 5980A mass spectrometer. Elemental analyses were performed by MHW Laboratories, Phoenix, Arizona.

9-Methylphenanthrene (**2**).

n-Butyllithium (170 ml of 1.6 *M* in hexane solution) was added *via* a syringe over a period of 35 minutes to a dry mixture of 9-bromophenanthrene (**1**) (27 g, 0.107 mole) and 400 ml of dry ether under a dry nitrogen stream at −78°. The mixture was allowed to warm slowly to room temperature and it was then stirred for 2 hours. The reaction mixture was cooled to −60 to −50° and dimethylsulfate (17.6 g, 0.139 mole) in 100 ml of

dry ether was added slowly. The reaction mixture was allowed to warm to room temperature and then refluxed for 8 hours. After cooling the reaction mixture was poured into a 15% aqueous solution of hydrochloric acid, extracted twice with benzene, dried over magnesium sulfate and evaporated *in vacuo* affording yellow crystals. Purification was accomplished by recrystallizing in hexane:benzene (2:1) affording colorless needles in 79% yield (16.3 g, 0.085 mole), mp 88-89° (lit [22] mp 90-91°).

9-Bromomethylphenanthrene (**3**).

A mixture of *N*-bromosuccinimide (15.9 g, 0.089 mole), 9-methylphenanthrene (**2**) (17 g, 0.088 mole), benzoyl peroxide (0.01 g) in dry benzene (250 ml) was gently refluxed overnight. The reaction mixture was cooled in an ice-bath and succinimide was separated by filtration. The last traces of imide were removed by shaking the filtrate with an ice cooled sodium hydroxide (5%) solution followed by shaking with water. The organic layer was dried over sodium sulfate, evaporated *in vacuo* affording a pale yellow oil in 96% yield (23 g, 0.085 mole); nmr (deuteriochloroform): 4.96 (s, 2H, CH_2-Br), 7.23-8.18 (m, H-1, H-2, H-3, H-6, H-7, H-8, H-10, 7H, ArH), 8.34-8.83 (m, 2H, H-4, H-5). This compound was not purified further but used in the next step (lit [24] mp 112-114°).

Diethyl 9-Phenanthrylmethylphosphonate (**4**).

A mixture of 9-bromomethylphenanthrene (**3**) (28 g, 0.10 mole) and triethyl phosphite (16.6 g, 0.10 mole) was heated at 150° for 8 hours giving upon distillation a pale yellow oil in 92% yield (30 g, 0.09 mole); nmr (deuteriochloroform): 1.13 (t, 6H, J = 6 Hz, 2 × CH_3), 3.57 (d, 2H, J = 20 Hz, CH_2-P), 3.91 (dq, J_{HH} = 8 Hz, J_{HP} = 8 Hz, 2 × CH_2CH_3), 7.31-7.88 (m, H-1, H-2, H-3, H-6, H-7, H-10, 6H, Ar*H*), 8.01-8.28 (dd, H-8, 1H, J = 4, 7 Hz), 8.4-8.8 (m, H-4, H-5, 2H); ms: m/e 328 (M^+, 100).

Anal. Calcd. for $C_{19}H_{21}O_3P$: C, 69.50; H, 6.45; O, 14.62. Found; C, 69.39; H, 6.40; O, 14.83.

1-(3'-Benzo[*b*]thienyl)-2-(9''-phenanthryl)ethene (**8**). Method A.

Sodium hydride (50% dispersion in mineral oil, 0.3 g, 0.012 mole) was placed in dry 1,2-dimethoxyethane (80 ml). The sodium hydride was used after washing twice with mixed hexanes solvent (50 ml). The slurry was cooled to 20° and diethyl 9-phenanthrylmethylphosphonate (**4**) (1.0 g, 0.03 mole) was added with stirring under a stream of nitrogen. After the addition, the solution was stirred at room temperature for 20 minutes. To the pale yellow solution maintained below 25°, benzo[*b*]thiophene-3-carboxaldehyde (**5**) (0.5 g, 0.03 mole) in 40 ml of dry 1,2-dimethoxyethane was slowly added. The solution was stirred at room temperature for 3.5 hours. The reaction mixture was slowly poured into a large excess of ice-water under a stream of nitrogen and the resulting precipitate was collected by filtration. The product was recrystallized in hexane:benzene (1:1) affording pale yellow needles in 69% yield (7.0 g, 0.021 mole), mp 162-163°.

Method B.

Compound **8** was prepared from diethyl 3-benzo[*b*]thenylphosphonate (**7**) (5.04 g, 0.018 mole) [3] and phenanthrene-9-carboxaldehyde (**6**) (4.1 g, 0.02 mole) in a manner similar to the preparation of Method A to give 2.7 g (0.08 mole, 45%) as pale yellow needles, mp 162-163°; nmr (deuteriochloroform): 7.16-8.30 (m, 2 × ethylene-*H*, aromatic-*H* of benzo[*b*]thiophene, H-1, H-2, H-3, H-6, H-7, H-8, H-10 of phenanthrene, 14H), 8.41-8.88 (m, H-4, H-5 of phenanthrene, 2H, Ar*H*); ms: m/e 336 (M^+, 100).

Anal. Calcd. for $C_{24}H_{16}S$: C, 85.68; H, 4.79; S, 9.53. Found: C, 85.41; H, 4.65; S, 9.40.

Benzo[*b*]triphenyleno[2,1-*d*]thiophene (**9**).

A solution of 1-(3'-benzo[*b*]thienyl)-2-(9''-phenanthryl)ethene (**8**) (1.0 g, 0.03 mole) and iodine (0.1 g) in dry benzene (360 ml) was irradiated for four hours with a 450 watt Hanovia medium pressure mercury lamp. To maximize the yields the whole amount of iodine was not added in the photocyclization vessel at once, but by adding small amounts of iodine (0.033 g) every hour for four hours. During the course of the reaction a slow stream of air was passed through the solution. The solvent was evaporated *in vacuo* and the residue (dark crystals) was chromatographed on a neutral alumina column using hexane:benzene (1:1) as the eluent affording colorless plates in 73% yield (0.73 g, 0.0022 mole), mp 180-181°; nmr (deuteriochloroform): 7.02-8.12 (m, H-2, H-3, H-6, H-7, H-10, H-11, H-12, H-13, H-14, 9H, Ar*H*), 8.29-8.95 (m, H-1, H-4, H-5, H-8, H-9, 5H, ArH); ms: m/e 334 (M^+, 100).

Anal. Calcd. for $C_{24}H_{14}S$: C, 86.19; H, 4.22; S, 9.59. Found: C, 86.24; H, 4.31; S, 9.40.

1-(2'-Benzo[*b*]thienyl)-2-(9''-phenanthryl)ethene (**12**). Method A.

Compound **12** was synthesized from the condensation of diethyl 9-phenanthrylmethylphosphonate (**4**) (2.5 g, 0.008 mole) and benzo[*b*]-thiophene-2-carboxaldehyde (**10**) (1.23 g, 0.008 mole) in a manner similar to the preparation of **8** and pale yellow prisms were obtained in 72% yield (1.8 g, 0.006 mole), mp 126-127°.

Method B.

This compound was prepared from the Wadsworth-Emmons reaction of phenanthrene-9-carboxaldehyde (**6**) (4.12 g, 0.020 mole) with diethyl 2-benzo[*b*]thenylphosphonate (**11**) (5.04 g, 0.0177 mole) [11] in a similar manner to the synthesis of **8** and pale yellow prisms were obtained in 35% yield (2.0 g, 0.006 mole), mp 126-127°; nmr (deuteriochloroform): 7.16-7.92 (m, 2 × ethylene-*H*, aromatic-*H* of benzo[*b*]thiophene, H-1, H-2, H-3, H-6, H-7, 12H), 7.90 (s, H-10 of phenanthrene, 1H, Ar*H*), 8.06-8.42 (m, H-8 of phenanthrene, 1H, Ar*H*), 8.50-8.92 (m, H-4, H-5 of phenanthrene, 2H, Ar*H*); ms: m/e 334 (M^+, 100).

Anal. Calcd. for $C_{24}H_{14}S$: C, 86.19; H, 4.22; S, 9.59. Found: C, 86.23; H, 4.01; S, 9.49.

Benzo[*b*]triphenyleno[1,2-*d*]thiophene (**13**).

Compound **13** was prepared by photocyclizing 1-(2'-benzo[*b*]thienyl)-2-(9''-phenanthryl)ethene (**12**) (1.0 g, 0.003 mole) in a manner similar to the preparation of **9**. The crude compound was chromatographed on a neutral alumina column using hexane:benzene (1:2) as the eluent affording colorless needles in 50% yield (0.5 g, 0.0015 mole), mp 151-152°; nmr (deuteriochloroform): 7.12-8.01 (m, H-2, H-3, H-6, H-7, H-10, H-12, H-13, H-14, 8H, Ar*H*), 8.28-8.91 (m, H-1, H-4, H-5, H-8, H-9, H-15, 6H, Ar*H*); ms: m/e 334 (M^+, 100).

Anal. Calcd. for $C_{24}H_{14}S$: C, 86.19; H, 4.22; S, 9.59. Found: C, 86.23; H, 4.01; S, 9.49.

1-Methyl-1-(2'-phenanthryl)-2-(2''-thienyl)ethene (**16**).

This compound was prepared from 2-acetylphenanthrene (**14**) (2.64 g, 0.012 mole) and diethyl 2-thenylphosphonate (**15**) (2.8 g, 0.012 mole) [2] in a manner similar to the preparation of **8** and the product was obtained. Purification was accomplished by column chromatography (neutral alumina) using hexane:benzene (1:2) as the eluent affording pale yellow needles in 50% yield (1.8 g, 0.006 mole), mp 88-89°; nmr (deuteriochloroform): 2.27 (s, CH_3 for *E* isomer), 2.51 (s, CH_3 for *Z* isomer) (*EZ* ratio = 2:1), 6.58-7.98 (m, C-*H*, aromatic-*H* of thiophene, H-1, H-3, H-6, H-7 and H-8, H-9, H-10 of phenanthrene, 11H), 8.48-8.80 (m, H-4, H-5 of phenanthrene, 2H, Ar*H*); ms: m/e 300 (M^+, 100).

Anal. Calcd. for $C_{21}H_{16}S$: C, 83.96; H, 5.37; S, 10.67. Found: C, 83.83; H, 5.43; S, 10.41.

5-Methylbenz[7,8]anthra[2,1-*b*]thiophene (**17**) and 1-Methylchryseno-[3,4-*b*]thiophene (**18**).

The mixture of compounds **17** and **18** was prepared from compound **16** (1.0 g, 0.0029 mole) in a manner similar to the preparation of compound **9**. Separation was accomplished on a basic alumina column using hexane and cyclohexane as the eluents. Compound **17** eluted (hexane) first in pure form. The second fraction consisted of a mixture of **17** and **18**. The third fraction (cyclohexane) consisted of pure **18**. The second fraction was separated on a second basic alumina column using pentane as the eluent. Compound **17** eluted first again in pure form from pentane followed by pure **18**. The total yield from the two column chromatographic separation of **17** was 31% (0.31 g, 0.0009 mole) and it was obtained as colorless crystals. Compound **18** was obtained in 34% yield (0.34 g,

0.0010 mole) from the two column chromatographic separation as pale yellow needles.

Compound **17**.

The colorless crystals of **17** had mp 244-245°; nmr (deuteriochloroform): 2.78 (s, CH_3, 3H), 7.18-8.01 (m, 6H, Ar*H*), 8.16 (s, H-6, 1H, Ar*H*), 8.28-8.89 (m, 3H, Ar*H*), 9.02 (s, H-13, 1H, Ar*H*); ms: m/e 348 (M^+, 100).

Anal. Calcd. for $C_{21}H_{14}S$: C, 84.53; H, 4.73; S, 10.74. Found: C, 84.41; H, 4.83; S, 10.63.

Compound **18**.

The pale yellow needles of **18** had mp of 222-223°; nmr (deuteriochloroform): 2.8 (s, CH_3, 3H), 7.5-8.2 (m, H-4, H-8, H-9, H-10, H-11, H-12, H-13, 7H), 8.3 (s, H-2, 1H), 8.5-9.2 (m, H-5, H-6, H-7, 3H); ms: m/e 348 (M^+, 100).

Anal. Calcd. for $C_{21}H_{14}S$: C, 84.53; H, 4.73; S, 10.74. Found: C, 84.49; H, 4.81; S, 10.71.

1-(4'-Dibenzothienyl)-2-(9''-phenanthryl)ethene (**21**). Method A.

This compound was prepared from the condensation of diethyl 9-phenanthrylmethylphosphonate (**4**) (2.0 g, 0.006 mole) and dibenzothiophene-4-carboxaldehyde (**19**) (1.3 g, 0.006 mole) [24] in a manner similar to the preparation of **8** and pale yellow crystals were obtained in 52% yield (1.23 g, 0.0032 mole), mp 234-235°; nmr (deuteriochloroform): 7.09-8.35 (m, 2 × ethylene-*H*, aromatic-*H* of dibenzothiophenes, H-1, H-2, H-3, H-6, H-7, H-8, H-10 of phenanthrene, 16H), 8.58-8.89 (m, H-4, H-5 of phenanthrene, 2H, Ar*H*); ms: m/e 386 (M^+, 100).

Method B.

This compound was synthesized from compound **6** (0.82 g, 0.004 mole) and diethyl 4-dibenzothenylphosphonate (**20**) (1.2 g, 0.004 mole) [18] in a similar manner to the preparation of **8** and pale yellow crystals were obtained in 64% yield (0.98 g, 0.0025 mole), mp 234-235°.

Anal. Calcd. for $C_{28}H_{18}S$: C, 87.01; H, 4.69; S, 8.30. Found: C, 86.86; H, 4.49; S, 8.58.

Triphenyleno[1,2-*c*]dibenzothiophene (**22**).

When compound **21** (1.0 g, 0.0026 mole) was photocyclized in a manner similar to the preparation of **9**, it gave **22** as colorless needles in 78% yield (0.78 g, 0.002 mole), mp 275-276°; nmr (deuteriochloroform): 7.18-8.28 (m, H-2, H-3, H-6, H-7, H-10, H-11, H-12, H-13, 8H, Ar*H*), 8.35-9.02 (m, H-1, H-4, H-5, H-8, H-9, H-14, H-16, H-17, 8H, Ar*H*); ms: m/e 384 (M^+, 100).

Anal. Calcd. for $C_{28}H_{16}S$: C, 87.47; H, 4.19; S, 8.34. Found; C, 87.32; H, 4.29; S, 8.01.

1-(1'-Dibenzothienyl)-2-(9''-phenanthryl)ethene (**25**). Method A.

This compound was prepared from diethyl 9-phenanthrylmethylphosphonate (**4**) (2.0 g, 0.006 mole) and dibenzothiophene-1-carboxaldehyde (**23**) (1.3 g, 0.0006 mole) [18] in a manner similar to the preparation of **8** affording pale yellow crystals in 60% yield (1.4 g, 0.003 mole), mp 216-217°.

Method B.

Compound **25** was synthesized from **6** (0.82 g, 0.004 mole) and diethyl 1-dibenzothenylphosphonate (**24**) (1.2 g, 0.004 mole) [18] in a manner similar to the preparation of **8** and pale yellow crystals were obtained in 57% yield (0.87 g, 0.0023 mole), mp 216-217°; nmr (deuteriochloroform): 7.12-8.45 (m, 2 × ethylene-*H*, aromatic-*H* of dibenzothiophene, H-1, H-2, H-3, H-6, H-7, H-8, H-10 of phenanthrene, 16H, Ar*H*); ms: m/e 386 (M^+, 100).

Anal. Calcd. for $C_{28}H_{18}S$: C, 87.01; H, 4.69; S, 8.30. Found: C, 86.90; H, 4.79; S, 8.53.

Triphenyleno[2,1-*a*]dibenzothiophene (**26**).

Compound **26** was obtained by photocyclizing **25** (1.0 g, 0.0029 mole) in a manner similar to **9** and colorless needles were obtained in 83% yield (0.93 g, 0.0024 mole), mp 218-219°; nmr (deuteriochloroform): 7.11-8.31 (m, H-2, H-3, H-6, H-7, H-10, H-12, H-14, 8H, Ar*H*), 8.40-9.31 (m, H-1, H-4, H-5, H-8, H-9, H-15, H-16, H-17, 8H, Ar*H*); ms: m/e 384 (M^+, 100).

Anal. Calcd. for $C_{28}H_{16}S$: C, 87.47; H, 4.19; S, 8.34. Found; C, 87.31; H, 4.34; S, 8.49.

1-(2'-Dibenzothienyl)-2-(9''-phenanthryl)ethene (**28**).

This compound was prepared from the phosphonate **4** (2.8 g, 0.0085 mole) and dibenzothiophene-2-carboxaldehyde (**27**) (1.8 g, 0.0085 mole) [24] and pale yellow needles were obtained in 80% yield (2.64 g, 0.0068 mole), mp 172-173°; nmr (deuteriochloroform): 7.15-8.30 (m, 2 × ethylene-*H*, aromatic-*H* of dibenzothiophene, H-1, H-2, H-3, H-6, H-7, H-8, H-10 of phenanthrene, 16H, Ar*H*), 8.34-8.92 (m, H-4, H-5 of phenanthrene, 2H, Ar*H*); ms: m/e 386 (M^+, 100).

Anal. Calcd. for $C_{28}H_{18}S$: C, 87.01; H, 4.69; S, 8.30. Found: C, 87.30; H, 4.73; S, 8.26.

Triphenyleno[1,2-*a*]dibenzothiophene (**29**) and Triphenyleno[2,1-*b*]dibenzothiophene (**30**).

The mixture was formed from the photocyclization of **28** (1.1 g, 0.0028 mole) in a manner similar to the preparation of **9**. The dark residue was chromatographed on a neutral alumina column eluting first with hexane to yield **29** and then with benzene:hexane (1:3) to yield **30**.

Compound **29**.

This compound eluted first and was obtained as colorless crystals (0.20 g, 0.00052 mole, 18%), mp 228°; nmr (deuteriochloroform): 7.3-7.6 (m, H-2 to H-15, 13H), 7.65 (dd, H-16, 1H, J = 7 Hz), 7.74 (dd, H-1, 1H, J = 7, 4 Hz); ms: m/e 384 (M^+, 100).

Anal. Calcd. for $C_{28}H_{16}S$: C, 87.47; H, 4.19; S, 8.34. Found: C, 87.40; H, 4.21; S, 8.39.

Compound **30**.

This compound eluted with benzene:hexane (1:3) as pale yellow needles (0.1 g, 0.00026 mole, 10%), mp 231°; nmr (deuteriochloroform): 7.3-8.4 (m, 11H), 8.4-8.6 (m, H-4, H-5, H-8, 3H), 8.65 (dd, H-1, 1H, J = 7, 3 Hz), 9.3 (s, H-15, 1H); ms: m/e (M^+, 100).

Anal. Calcd. for $C_{28}H_{16}S$: C, 87.47; H, 4.19; S, 8.34. Found; C, 87.56; H, 4.21; S, 8.08.

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