C_2Cl_4)[19] 1756 (s), 1729 (s), 1407, 1281 (m), 1199 (s), 1177, 1136 (s), 1123 (sh), 1092 (s), 995 (sh), 956, 865; MS *m/e* 342 (<1, M^+), 341 (1), 131 (100, $M - C_{15}H_{31}$), 103 (10, $M - C_{15}H_{31}CO$), 43 (49). Anal. Calcd for $C_{20}H_{38}O_4$: C, 70.13; H, 11.18; O, 18.69. Found: C, 69.97; H, 11.37; O, 18.51.

***trans*-3a:** yield 1.39 g (46%, based on **2a**); mp 30–30.5 °C; IR (CS_2, C_2Cl_4)[19] 1748 (s), 1726 (s), 1410, 1335, 1279 (m), 1199 (s), 1135 (s), 1120 (sh), 1094 (s), 1045 (m), 990, 947, 860; MS *m/e* 342 (<1), 341 (1), 131 (100), 103 (6), 43 (12). Anal. Found: C, 70.16; H, 11.17; O, 18.67.

***cis*-3b:** yield, 1.04 g (31%, based on **2b**); liquid at 0 °C; IR (liquid film)[19] 2940 (m), 1748 (s), 1723 (s), 1640, 1403 (m), 1340 (m), 1284 (m), 1202 (s), 1180 (sh), 1135 (s), 1092 (s), 1058 (m), 1028 (m), 960, 933 (m), 865; MS *m/e* 368 (1, M^+), 367 (1), 131 (100, $M - C_{17}H_{33}$), 103 (20, $M - C_{17}H_{33}CO$), 43 (56). Anal. Calcd for $C_{22}H_{40}O_4$: C, 71.70; H, 10.94; O, 17.36. Found: C, 71.74; H, 10.93; O, 17.21.

***trans*-3b:** yield 1.38 g (42%, based on **2b**); liquid at 0 °C; IR (liquid film)[19] 2958 (m), 1752 (s), 1730 (s), 1645, 1412 (m), 1342 (m), 1285 (m), 1205 (s), 1136 (s), 1095 (s), 1045 (m), 1023 (sh), 957 (sh), 938 (m), 867; MS *m/e* 368 (1), 367 (1), 131 (100), 103 (19), 43 (49). Anal. Found: C, 71.66; H, 10.95; O, 17.14.

2-Alkyl-4-hydroxymethyl-1,3-dioxolanes (4) were prepared from the four respective 4-methoxycarbonyl acetals (*cis*- and *trans*-**3a**, and *cis*- and *trans*-**3b**) by reduction in a saturated solution of $LiAlH_4$[5,10] in dry Et_2O (dropwise addition of **3**, reflux for 2 h, decomposition of excess $LiAlH_4$ with moist Et_2O) and extraction from the basic medium followed by TLC[7] purification (R_f 0.56; developing solvent, hexane–Et_2O, 40:60, v/v) produced the stereomeric five-ring glycerol acetals (**4a, 4b**) in essentially quantitative yields.

2-Pentadecyl-4-hydroxymethyl-1,3-dioxolanes (4a). *cis*-4a: mp 41.5–42.5 °C. Anal. Calcd for $C_{19}H_{38}O_3$: C, 72.56; H, 12.18; O, 15.26. Found: C, 72.71; H, 12.25; O, 15.47.

***trans*-4a:** mp 44.5–45.5 °C. Anal. Found: C, 72.44; H, 12.31; O, 15.32.

2-(*cis*-8′-Heptadecenyl)-4-hydroxymethyl-1,3-dioxolanes (4b). *cis*-4b: liquid at 0 °C. Anal. Calcd for $C_{21}H_{40}O_3$: C, 74.07; H, 11.84; O, 14.09. Found: C, 73.87; H, 11.73; O, 14.28.

***trans*-4b:** liquid at 0 °C. Anal. Found: C, 73.87; H, 11.69; O, 14.44.

2-Alkyl-4-acetoxymethyl-1,3-dioxolanes (5) were prepared from the individual hydroxymethyl acetals 4 by acetylation with 100 parts (v/w) of acetic anhydride in the presence of 10 parts (v/w) of dry pyridine for 2 h at 80 °C.[5] After extraction from the basic medium, the acetates were purified by TLC[7] (R_f 0.46; developing solvent, hexane–Et_2O, 70:30, v/v). All physical characteristics of the pentadecyl derivatives *cis*- and *trans*-**5a** were identical to those reported previously for the respective five-ring glycerol acetal acetates prepared by an alternate route.[5]

2-(*cis*-8′-Heptadecenyl)-4-acetoxymethyl-1,3-dioxolanes (5b). *cis*-5b: liquid at 0 °C; MS *m/e* 382 (1 M^+), 381 (1), 145 (100, $M - C_{17}H_{33}$), 117 (95, $M - C_{17}H_{33}CO$), 43 (74). Anal. Calcd for $C_{23}H_{42}O_4$: C, 72.21; H, 11.06; O, 16.73. Found: C, 71.99; H, 10.86; O, 17.15.

***trans*-5b:** liquid at 0 °C; MS, *m/e* 382 (1, M^+), 381 (2), 145 (100, $M - C_{17}H_{33}$), 117 (82, $M - C_{17}H_{33}CO$), 43 (84). Anal. Found: C, 71.99; H, 11.00; O, 17.00.

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Registry No.—**1,** 615-34-9; **2a,** 629-80-1; **2b,** 2423-10-1; glyceric acid calcium salt hydrate, 6057-35-8; ethanediol, 107-21-1.

References and Notes

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Synthesis of Alkyl-Substituted Benzo[*c*]phenanthrenes and Chrysenes by Photocyclization

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The synthesis of alkyl-substituted polycyclic aromatic hydrocarbons is often necessary to provide samples to aid trace analyses of these compounds in environmental samples. The reported syntheses of monomethylchrysenes and monomethylbenzo[*c*]phenanthrenes, many of which are reported to be carcinogenic,[1] are generally multistep.[2,3]

We report herein on the syntheses of alkyl-substituted benzo[*c*]phenanthrenes and chrysenes by photocyclization[4] of the requisite naphthylstyrenes.[5,6] Since naphthylstyrenes can be readily prepared via the Wittig or Grignard reactions, this procedure appeared to offer a convenient general synthetic route to alkylchrysenes and alkylbenzo[*c*]phenanthrenes.

The six isomeric monomethylbenzo[*c*]phenanthrenes **2a–f** were prepared as outlined in Scheme I, in yields ranging from 66 to 89% (Table I). The spectral properties of these compounds (^1H NMR, UV) correlate well with published data.[7,8]

In addition, ^1H NMR and GLC data indicated the photocyclization products were free of benzo[*c*]phenanthrene and other isomeric methylbenzo[*c*]phenanthrenes. In all preparations, however, we found small amounts (1%) of isomeric methylbenz[*a*]anthracenes produced through cyclization involving the β position of the naphthalene moiety. However, the chromatographic properties of the benzo[*c*]phenanthrenes and the benz[*a*]anthracenes on alumina permit facile sepa-

Table I. Benzo[*c*]phenanthrenes and Chrysenes Prepared by Photocyclization[a]

Compd	Registry no.	% yield[b]	Mp, °C	Lit. mp, °C
2a	4076-39-5	89	140–141.5	136.8–141[2c]
2b	2606-85-1	52	80–81	80.6–81.4[2c]
2c	2381-19-3	66	53–54.5	54.4–55.4[2c]
2d	4076-40-8	68	65–66	64.6–65.6[2c]
2e	652-04-0	72	69–70	70.6–71.6[2c]
2f	2381-34-2	72	76–77	76.8–77.6[2c]
2g	4176-45-8	94	91.5–92	90–91[2b]
4a	3697-24-3	65	116–117	116.8–117.6[13]
4b	54986-62-8	80	91–92	91.4–92.4[14]
6	202-98-2	8	171–173	172.4–172.9[15]

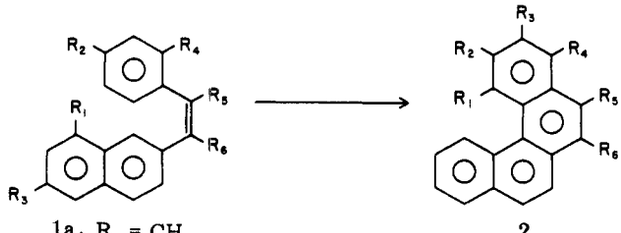
[a] See Experimental Section for general photocyclization procedure. [b] Isolated yields following column chromatography.

Table II. Preparation of Naphthylstyrenes Via the Wittig Reaction[a]

Napthyl-styrene[b]	Registry no.	Alkyl bromide[c]	Registry no.	Carbonyl Compd[c]	Registry no.	% yield[d]	Mp, °C	Lit. mp, °C
1a	63216-64-8	Benzyl bromide	100-39-0	1-Methyl-7-naphthaldehyde[a]	63216-67-1	94	80–81	
b	35160-96-4	4-Methylbenzyl bromide	104-81-4	2-Naphthaldehyde	66-99-9	88	189–190.5	(188–189)[16]
c	63216-65-9	2-Bromomethyl-6-methylnaphthalene[12]	52988-15-5	Benzaldehyde	100-52-7	73	173–174	
d	63216-66-0	2-Methylbenzyl bromide	89-92-9	2-Naphthaldehyde		87	86–87	
e	20883-24-3	2-Bromomethylnaphthalene	939-26-4	Acetophenone	98-86-2	59	146–147	(147.5–148)[17]
f	17181-02-1	Benzyl bromide		2′-Acetonaphthone	98-08-3	81	137–138	(139)[6b]
g	23833-60-5	2-Bromomethylnaphthalene		4-Methoxybenzaldehyde	123-11-5	96	135–136	(134–135)[18]
3a	63269-87-4	Benzyl bromide		1′-Acetonaphthone	941-98-0	81	oil	
b	63269-88-5	Benzyl bromide		1′-Propionaphthone	2876-63-3	84	oil	
5	21844-25-7	Benzyl bromide		Acenaphthone	2235-15-6	14	98–99	(99–100)[11]

[a] See Experimental Section for preparation. [b] Accurate mass measurements were obtained on all molecular ions. Experimental values were in agreement (±0.001 amu) with calculated values. [c] All compounds were obtained from Aldrich Chemical Co., Milwaukee, Wis., unless otherwise noted. [d] Isolated yields following column chromatography.

Scheme I



1a, $R_1 = CH_3$
b, $R_2 = CH_3$
c, $R_3 = CH_3$
d, $R_4 = CH_3$
e, $R_5 = CH_3$
f, $R_6 = CH_3$
g, $R_2 = OCH_3$

2

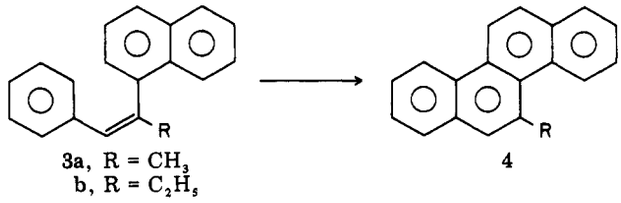
Note: R_n = H unless otherwise indicated.

rations of these isomers. The preparations of the naphthylstyrene precursors are documented in Table II.

The photocyclization of α-naphth-2-ylstyrenes, on the other hand, can yield only chrysenes (Scheme II). The yields are comparable to cyclizations involving the benzo[*c*]phenanthrenes (Table I). Again spectral properties (^{1}H NMR, UV) correlate well with previously published data.[1b,9,10]

Since the photocyclization procedure has been utilized in the preparation of phenanthrenes with fluoro, chloro, bromo, methoxyl, trifluoromethyl, carboxyl, phenyl, hydroxy and

Scheme II

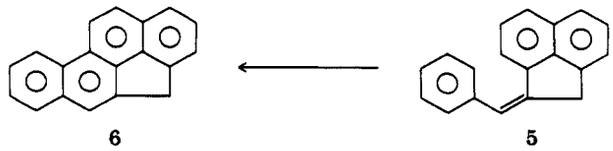


3a, $R = CH_3$
b, $R = C_2H_5$

4

cyano substituents,[6b] the synthesis by this route of similarly substituted benzo[*c*]phenanthrenes and chrysenes seemed feasible. Hence, 2-methoxybenzo[*c*]phenanthrene was prepared in good yield. However, 4,5-methylene chrysene (**6**), a reported carcinogen, was prepared in low yield, possibly due to the increased distance between the potential reactive centers in (**5**). Attempted synthesis of **5** from 1,8-dibromomethylnaphthalene by the method of Bestman et al.[11] produced only acenaphthene.

Scheme III



6 5

In conclusion, the photocyclization preparation of alkyl-substituted benzo[c]phenanthrenes and chrysenes offers a convenient, general synthetic route, with distinct advantages over previously published procedures.

Experimental Section

Melting points were obtained on a Thomas Hoover Uni Melt and are corrected. Microanalyses were performed by Micro-Tek Associates, Skokie, Ill. The IR, ^1H NMR, UV, and MS data were consistent with the assigned structures. The IR data were recorded on a Beckman IR-9, ^1H NMR data on Varian Associates Model HA-100 or CFT-20, UV data on a Cary 14 or Beckman Acta CIII, MS data on an AEI MS-9 equipped with a DS-30 data system. MS samples were introduced either via a variable temperature direct probe (Variset Co., Madison, Wis.) or a GC inlet.

1-Methyl-7-naphthaldehyde. To a stirred solution of 5.0 g (22 mmol) of 7-bromo-1-methylnaphthalene in 125 mL of dry ether was added 50 mmol of *N*-butyllithium in hexane (Alpha Chemical Co., Danvers, Mass.). After stirring for 30 min at room temperature, 7.8 mL (100 mmol) of dry DMF was added in one portion. Following 2 h of additional stirring, the solution was treated with 50 mL of 6 N HCl. The organic phase was washed with 50 mL of H_2O, dried (Na_2SO_4), and concentrated in vacuo. Recrystallization of the yellow solid from hexane gave 2.76 g (86%) of the aldehyde as white needles (mp 55.5–56.5 °C): IR ($\nu_{C=O}$) 1680 cm^{-1}; NMR ($CDCl_3$) s, δ 10.12 (1 H, CHO), m, 8.40–7.20 (6 H, aromatic), s, 2.72 (3 H, CH_3).

Anal. Calcd for $C_{12}H_{10}O$: C, 84.68, H, 5.92. Found: C, 84.71, H, 5.89.

2-Bromomethyl-6-methylnaphthalene. A solution of 20.25 g (0.13 mol) of 2,6-dimethylnaphthalene and 20.76 g (0.12 mol) of *N*-bromosuccinimide in 250 mL of carbon tetrachloride was refluxed under UV irradiation for 2 h in the presence of a catalytic amount of benzoyl peroxide. The reaction mixture was cooled and the succinimide removed by filtration. The solution was washed with sodium bisulfite, dried (Na_2SO_4), concentrated, and chromatographed on alumina, eluting with benzene/hexane to give 13.29 g (44%). Recrystallization from ethanol gave crystals: mp 160–161 °C (dec) (lit 92–93 °C);[12] NMR ($CDCl_3$) m, δ 7.78–7.12 (6 H, aromatic), s, 4.66 (2 H, CH_2Br), s, 2.45 (3 H, CH_3). Exact mass (M^+) calcd for $C_{12}H_{11}Br$: 234.0044. Found: 234.0056. Anal. Calcd for $C_{12}H_{11}Br$: C, 61.30; H, 4.72. Found: C, 61.23; H, 4.82.

General Synthetic Procedure for Preparation of the Naphthylstyrene via the Wittig Reaction. A solution of 125 mL of anhydrous DMF, 0.10 mol of the benzyl halide, and 0.10 mol of triphenylphosphine was stirred magnetically at reflux for 1.5 h in a 250-mL round-bottom flask fitted with a reflux condenser. The mixture was cooled to room temperature and the white precipitate was filtered, washed with ether, and dried overnight at 50 °C in vacuo (yields 82–97%).

A solution of 380 mL of freshly prepared 0.2 M sodium ethoxide (0.077 mol) in ethanol was added over 20 min to a stirred solution of 0.07 mol of a benzyltriphenylphosphonium bromide in 75 mL of dry ethanol at room temperature. The resultant ylide was stirred for 10 min and a solution was added consisting of 0.07 mol of the acetylnaphthalene in 25 mL of dry ethanol. After refluxing for 8 h, the milky white solution had turned bright yellow. The solvent was removed in vacuo, taken up in ether, washed with H_2O, concentrated, and chromatographed on alumina eluting with hexane. A mixture of the cis and trans isomers was usually obtained.

General Synthetic Procedure for the Preparation of Alkylbenzo[c]phenanthrenes and Chrysenes via Photocyclization. A solution of 0.01 mol of the appropriate naphthylstyrene and 127 mg of iodine in 500 mL of freshly distilled cyclohexane was placed in a 500-mL quartz tube equipped with a gas-dispersion tube at the bottom. Irradiation for 12 h (Rayonet Preparative Photochemical Reactor RPR-208, New South England Ultraviolet Co., Middleton, Conn.) with 3000-Å lamps and a brisk air flow through the dispersion tube resulted, in most instances, in precipitation of solid material. The solid was dissolved in chloroform, combined with the cyclohexane supernatant, and concentrated in vacuo. The residue was absorbed into 1.5 g of neutral alumina, placed on a 2× 150-mm column of the same, and eluted with cyclohexane to yield the desired alkylpolycyclic aromatic hydrocarbon.

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Registry No. —Triphenylphosphine, 603-35-0; 7-bromo-1-methylnaphthalene, 33295-35-1; 2,16-dimethylnaphthalene, 581-42-0.

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