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Synthesis and Fluorescence Behaviour of Some 3-Cyano-4-Substituted-6-Pyrenyl-2-Pyridone Derivatives

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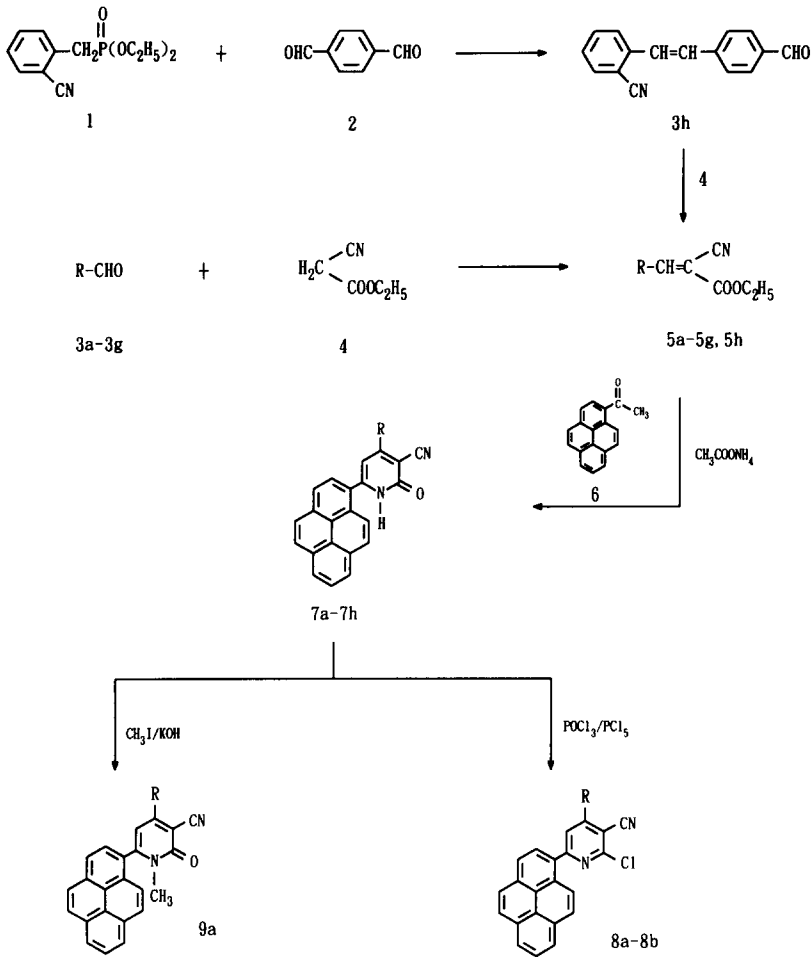
ABSTRACT

*A series of 3-cyano-4-substituted-6-pyrenyl-2-pyridone derivatives (**7a–7h**) was prepared by cyclization of ethyl 2-cyano-3-substituted acrylates (**5a–5h**) with 3-acetylpyrene (**6**) in the presence of ammonium acetate. Reaction of compounds **7a** and **7b** with $POCl_3/PCl_5$ gave the 2-chloro-3-cyano-4-substituted-6-pyrenylpyridines (**8a** and **8b**). 1-Methyl-3-cyano-4-phenyl-6-pyrenyl-2-pyridone (**9a**) was prepared by interaction of compound **7a** and methyl iodide. The structures of the compounds were confirmed by IR, ^1H NMR, mass spectra and elemental analysis. The fluorescence behaviour of compounds **7a–7h** was also studied*

1 INTRODUCTION

In a previous paper,[1] we reported the synthesis of 6-pyrenylpyridine derivatives which were shown to exhibit blue to green fluorescence emission. The synthetic methods for various substituted 2-pyridones have been reported,[2–7] and such compounds have widespread use in the synthesis of dyes,[8–11] fluorescent dyes,[12] thermal transfer recording materials,[13] and electron conductive materials,[14] etc. We report here the preparation and fluorescent properties of some 6-pyrenyl-2-pyridone derivatives; these compounds were synthesized according to the method for 2-pyridone derivatives described by Kambe *et al.*[15]



Compound					R
3a	5a	7a	8a	9a	Phenyl
3b	5b	7b	8b		4-Methoxyphenyl
3c	5c	7c			4-Methylphenyl
3d	5d	7d			4-Cyanophenyl
3e	5e	7e			Styryl
3f	5f	7f			1-Naphthyl
3g	5g	7g			2-Thienyl
3h	5h	7h			4-(2-Cyanostyryl)benzyl

Scheme 1

2 RESULTS AND DISCUSSION

2.1 Synthesis

The routes used to prepare the 3-cyano-4-substituted-6-pyrenyl-2-pyridones (**7a–7h**), 2-chloro-3-cyano-4-substituted-6-pyrenylpyridines (**8a** and **8b**) and 1-methyl-3-cyano-4-phenyl-6-pyrenyl-2-pyridone (**9a**) are shown in Scheme 1.

Valenti[16] reported the synthesis of 4-(2-cyanostyryl)benzaldehyde (**3h**) by oxidation processes and in this present paper, **3h** was obtained using the Wittig reaction.[17,18] This process is best carried out by dissolving the terephthalaldehyde (**2**) in ethyl acetate and then adding the diethyl *o*-cyano-benzylphosphonate (**1**), followed by slow addition of the necessary amount of alkali required for the condensation. The ethyl 2-cyano-3-substituted acrylates (**5a–5h**) were prepared by condensation of ethyl cyanoacetate (**4**) with aromatic and heteroaromatic aldehydes (**3a–3h**) in ethanol and they were then cyclized with compound **6** in the presence of

TABLE 1
IR and ^{1}H NMR Data for 4-(2-Cyanostyryl)benzaldehyde (**3h**) and Ethyl 2-Cyano-3-Substituted Acrylates (**5a–5d**)

Compound no.	*IR: ν KBr (cm^{-1})*	*^{1}H NMR: δ (ppm) ($CDCl_3$)*
3h	1691 [—C=O]	7·23–7·88 [m, 10H, 4-(2-cyanostyryl)benzyl]
	2219 [—C≡N]	9·99 [s, 1H, CHO on phenyl]
5a	1726 [—C=O]	1·35–1·39 [t, 3H, —$CH_2\underline{CH_3}$]
	2225 [—C≡N]	4·32–4·39 [q, 2H, —$\underline{CH_2}CH_3$]
		7·45–7·53 [m, 5H, phenyl]
		8·22 [s, 1H, —$\underline{CH}$=C—]
5b	1722 [—C=O]	1·32–1·36 [t, 3H, —$CH_2\underline{CH_3}$]
	2216 [—C≡N]	3·84 [s, 3H, OCH_3 on phenyl]
		4·28–4·35 [q, 3H, —$\underline{CH_2}CH_3$]
		6·93–7·96 [dd, 4H, 2,6- and 3,5-position on phenyl]
		8·11 [s, 1H, —$\underline{CH}$=C—]
5c	1723 [—C=O]	1·34–1·38 [t, 3H, —$CH_2\underline{CH_3}$]
	2217 [—C≡N]	2·39 [s, 3H, CH_3 on phenyl]
		4·30–4·38 [q, 3H, —$\underline{CH_2}CH_3$]
		7·25–7·87 [dd, 4H, 2,6- and 3,5-position on phenyl]
		8·17 [s, 1H, —$\underline{CH}$=C—]
5d	1738 [—C=O]	1·36–1·40 [t, 3H, —$CH_2\underline{CH_3}$]
	2227 [—C≡N]	4·34–4·42 [q, 3H, —$\underline{CH_2}CH_3$]
		7·75–8·04 [dd, 4H, 2,6- and 3,5-position on phenyl]
		8·21 [s, 1H, —$\underline{CH}$=C—]

excess ammonium acetate to give the 3-cyano-4-substituted-6-pyrenyl-2-pyridones (**7a–7h**).

Following established procedures in pyridine chemistry,[19–22] compounds **7a** and **7b** were treated with $POCl_3$ and PCl_5 to give 2-chloro-6-pyridines (**8a** and **8b**). Bomika *et al.*[23] have reported the alkylation of 2-pyridone at the 1-position. Reaction of 3-cyano-4-phenyl-6-pyrenyl-2-pyridone (**7a**) and methyl iodide in KOH–ethanol gave 1-methyl-3-cyano-4-phenyl-6-pyrenyl-2-pyridone (**9a**).

IR and ^1H NMR characterization data for the compounds synthesized are shown in Tables 1–4.

2.2 Electron spectra

The absorption and fluorescence emission maxima of the 3-cyano-4-substituted-6-phenyl-2-pyridones (**7a–7h**) in DMF are listed in Table 5. Relative to compound **7a**, the presence of a methoxy group (in **7b**) or a methyl group (in **7c**) on the phenyl ring, or of a naphthyl group (in **7g**), at the 4-position of the pyridone ring, results in hypsochromic shifts, but introduction of a cyano group (in **7d**), a styryl group (in **7e**) or stilbene residue (in **7h**) has the reverse effect. Replacement of the phenyl ring in **7a**

TABLE 2
IR and ^1H NMR Data for Ethyl 2-Cyano-3-Substituted Acrylates (**5e–5h**)

Compound no.	*IR: ν KBr (cm^{-1})*	*^1H NMR: δ (ppm) ($CDCl_3$)*
5e	1722 [—C=O]	1·32–1·37 [t, 3H, —$CH_2\underline{CH_3}$]
	2222 [—C≡N]	4·28–4·35 [q, 2H, —$\underline{CH_2}CH_3$]
		7·24–7·57 [m, 7H, phenyl and —$\underline{CH}$=CH—$\underline{CH}$—]
		7·95–7·99 [q, 1H, —CH=$\underline{CH}$—CH=]
5f	1723 [—C=O]	1·39–1·44 [t, 3H, —$CH_2\underline{CH_3}$]
	2223 [—C≡N]	4·38–4·45 [q, 3H, —$\underline{CH_2}CH_3$]
		7·58–8·30 [m, 7H, naphthyl]
		9·07 [s, 1H, —$\underline{CH}$=C—]
5g	1722 [—C=O]	1·32–1·37 [t, 3H, —$CH_2\underline{CH_3}$]
	2218 [—C≡N]	4·29–4·36 [q, 3H, —$\underline{CH_2}CH_3$]
		7·17–7·20 [q, 1H, 4-position on thienyl]
		7·74–7·80 [dd, 2H, 3,5-position on thienyl]
		8·30 [s, 1H, —$\underline{CH}$=C—]
5h	1722 [—C=O]	1·36–1·41 [t, 3H, —$CH_2\underline{CH_3}$]
	2223 [—C≡N]	4·33–4·41 [q, 2H, —$\underline{CH_2}CH_3$]
		7·24–7·81 [m, 8H, 2 phenyl on 4-(2-cyanostyryl)benzyl]
		7·99–8·02 [d, 2H, —$\underline{CH}$=$\underline{CH}$—]
		8·21 [s, 1H, —$\underline{CH}$=C—]

TABLE 3
IR and ^{1}H NMR Data for 3-Cyano-4-Substituted-6-Pyrenyl-2-Pyridones (**7a–7f**)

Compound no.	*IR: ν KBr (cm^{-1})*	*^{1}H NMR: δ (ppm) (DMSO-d_6)*	
7a	1658 [—C=O]	6·72	[s, 1H, 5-position on pyridone]
	2221 [—C≡N]	7·55–7·79	[m, 5H, phenyl]
		8·11–8·41	[m, 9H, pyrenyl]
		13·12	[br, 1H, OH on pyridone]
7b	1643 [—C=O]	3·83	[s, 3H, OCH_3 on phenyl]
	2221 [—C≡N]	6·70	[s, 1H, 5-position on pyridone]
		7·09–7·78	[dd, 4H, 2,6- and 3,5-position on phenyl]
		8·11–8·41	[m, 9H, pyrenyl]
		13·02	[br, 1H, OH on pyridone]
7c	1644 [—C=O]	2·37	[s, 3H, CH_3 on phenyl]
	2222 [—C≡N]	6·68	[s, 1H, 5-position on pyridone]
		7·34–7·68	[dd, 4H, 2,6- and 3,5-position on phenyl]
		8·10–8·40	[m, 9H, pyrenyl]
		13·07	[br, 1H, OH on pyridone]
7d	1650 [—C=O]	6·78	[s, 1H, 5-position on pyridone]
	2226 [—C≡N]	7·95–8·42	[m, 13H, 2,6- and 3,5-position on phenyl and pyrenyl]
		13·25	[br, 1H, OH on pyridone]
7e	1646 [—C=O]	7·14	[s, 1H, 5-position on pyridone]
	2220 [—C≡N]	7·29–7·34	[d, 1H, —CH=CH—]
		7·38–7·68	[m, 5H, phenyl]
		7·86–7·91	[d, 1H, —CH=CH—]
		8·11–8·43	[m, 9H, pyrenyl]
		12·81	[br, 1H, OH on pyridone]
7f	1633 [—C=O]	6·63	[s, 1H, 5-position on pyridone]
	2222 [—C≡N]	7·60–8·39	[m, 16H, naphthyl and pyrenyl]
		13·23	[br, 1H, OH on pyridone]

by a heteroaryl ring (**7g**) causes a bathochromic shift of 7 nm. The chloro derivatives (**8a** and **8b**) and the *N*-alkylated compound (**9a**) show hypsochromic shifts of 8–18 nm. The fluorescence emission maxima of compounds **7a–7c** and **7f** are in the range 483–486 nm and these compounds show a strong bluish-green fluorescence in DMF. The emission maxima of compounds **7d**, **7e** and **7h**, and of the heteroaryl compound (**7g**) were at longer wavelength. Compound **7e**, containing a styryl residue on the pyridone ring, has the largest fluorescence emission maximum at 514 nm, but shows only a weak green fluorescence. The chloro-containing compounds (**8a** and **8b**), and the *N*-methyl derivative (**9a**) show bathochromic shifts of 4–9 nm in the fluorescence maxima.

TABLE 4

IR and ^{1}H NMR Data for 3-Cyano-4-Substituted-6-Pyrenyl-2-Pyridones (**7g** and **7h**), 2-Chloro-3-Cyano-4-Substituted-6-Pyrenylpyridines (**8a** and **8b**) and 1-Methyl-3-Cyano-4-Phenyl-6-Pyrenyl-2-Pyridone (**9a**)

Compound no.	*IR: ν KBr (cm^{-1})*	*^{1}H NMR: δ (ppm) (DMSO-d_6)*	
7g	1629 [—C=O]	6·87	[s, 1H, 5-position on pyridone]
	2211 [—C≡N]	7·29–7·32	[q, 1H, 4-position on thienyl]
		7·98–8·41	[m, 11H, 3,5-position on thienyl and pyrenyl]
		13·02	[br, 1H, OH on pyridone]
7h	1642 [—C=O]	6·76	[s, 1H, 5-position on pyridone]
	2219 [—C≡N]	7·45–8·01	[m, 10H, 4-(2-cyanostyryl)benzyl]
		8·11–8·42	[m, 9H, pyrenyl]
		13·11	[br, 1H, OH on pyridone]
8a	2225 [—C≡N]	7·61–7·87	[m, 5H, phenyl]
		8·11	[s, 1H, 5-position on pyridine]
		8·13–8·53	[m, 9H, pyrenyl]
8b	2223 [—C≡N]	3·85	[s, 3H, OCH_3 on phenyl]
		7·14–7·84	[dd, 4H, 2,6- and 3,5-position on phenyl]
		8·05	[s, 1H, 5-position on pyridine]
		8·10–8·51	[m, 9H, pyrenyl]
9a	1632 [—C=O]	3·31	[s, 3H, N—CH_3]
	2220 [—C≡N]	6·54	[s, 1H, 5-position on pyridone]
		7·47–7·77	[m, 5H, phenyl]
		7·91–8·30	[m, 9H, pyrenyl] ($CDCl_3$)

TABLE 5

Absorption and Fluorescence Spectral Data for 6-Pyrenyl-2-Pyridones (**7a–7h**), 2-Chloro-6-Pyrenylpyridines (**8a** and **8b**) and 1-Methyl-6-Pyrenyl-2-Pyridone (**9a**) in Dimethylformamide

Compound no.	*λ_{max} (nm)*	*Log ε*	*λ_{em} (nm)*
7a	382	4·51	486
7b	380	4·33	483
7c	380	4·40	486
7d	388	4·26	505
7e	388	4·58	514
7f	372	4·29	485
7g	389	4·30	503
7h	385	4·35	509
8a	374	4·39	490
8b	373	4·37	487
9a	364	4·39	495

3 EXPERIMENTAL

All melting points are uncorrected. IR spectra (KBr) were recorded on a JASCO Hc-2 FT-IR/IR-3. The ^{1}H NMR spectra were determined with a JUM-FX-100 (JEOL) FT-NMR spectrometer using TMS as internal standard. The mass spectra were determined on a FINNIGAN TSQ-700 mass spectrometer. Absorption spectra were recorded on a Milton Roy UV-1201 recording spectrophotometer and the concentration of solution used for the measurements was 5×10^{-5} M. Fluorescence spectra were recorded on a Perkin-Elmer LS 50 luminescence spectrometer; the concentration of the solution used for these measurements was 5×10^{-8} M.

3.1 4-(2-cyanostyryl)benzaldehyde (3h)

30% sodium methylate solution in methanol (20 g) was added dropwise over 5 h to a mixture of *o*-cyanobenzylphosphonate (13·6 g, 0·05 mole) and terephthalaldehyde (6·7 g, 0·05 mole) in ethyl acetate (200 g) at reflux, displacing the air by nitrogen. After the reaction, the resulting solution was cooled and the product filtered and washed with methanol to give pale yellow crystals (60%), m.p. 146–147°C (P^+ at *m/e* 233). Calculated for $C_{16}H_{11}NO$: C, 82·40; H, 4·72; N, 6·01. Found: C, 82·35; H, 4·71; N, 5·94.

3.2 Ethyl 2-cyano-3-phenylacrylate (5a)

A mixture of ethyl cyanoacetate (2·26 g, 0·02 mole), benzaldehyde (2·12 g, 0·02 mole), ethanol (50 ml) and piperidine (1 ml) was stirred at room temperature overnight. Alcohol was removed on a rotary evaporator and the residue poured into ice to give a viscous product which was recrystallized from ethanol to give white crystals (83%), m.p. 45–48°C (P^+ at *m/e* 201). Calculated for $C_{12}H_{11}NO_2$: C, 71·61; H, 5·51; N, 6·96. Found: C, 71·51; H, 5·59; N, 6·91.

3.3 Ethyl 2-cyano-3-(4-methoxyphenyl) acrylate (5b)

Compound **5b** was prepared in a manner similar to that described above for **5a**. It was recrystallized from ethanol as pale yellow crystals (93%), m.p. 78–81°C (P^+ at *m/e* 231). Calculated for $C_{13}H_{13}NO_3$: C, 67·51; H, 5·67; N, 6·06. Found: C, 67·23; H, 5·47; N, 6·26.

3.4 Ethyl 2-cyano-3-(4-methylphenyl) acrylate (5c)

Compound **5c** was prepared in a manner similar to that described above for **5a**. During the reaction, the white product precipitated. The crude product

was filtered and recrystallized from ethanol to give white crystals (80%), m.p. 92–94°C (P^+ at *m/e* 215). Calculated for $C_{13}H_{13}NO_2$: C, 72·56; H, 6·05; N, 6·51. Found: C, 72·64; H, 6·05; N, 6·24.

3.5 Ethyl 2-cyano-3-(4-cyanophenyl) acrylate (5d)

Compound **5d** was prepared in a manner similar to that described above for **5c**. It was recrystallized from ethanol as white crystals (93%), m.p. 167–170°C (P^+ at *m/e* 226). Calculated for $C_{13}H_{10}N_2O_2$: C, 69·00; H, 4·46; N, 12·39. Found: C, 68·98; H, 4·21; N, 12·38.

3.6 Ethyl 2-cyano-3-styryl acrylate (5e)

Compound **5e** was prepared in a manner similar to that described above for **5c**. It was recrystallized from ethanol as pale yellow crystals (70%), m.p. 96–100°C (P^+ at *m/e* 227). Calculated for $C_{14}H_{13}NO_2$: C, 74·01; H, 5·73; N, 6·17. Found: C, 74·01; H, 5·67; N, 6·11.

3.7 Ethyl 2-cyano-3-(1-naphthyl) acrylate (5f)

Compound **5f** was prepared in a manner similar to that described above for **5a**. It was recrystallized from ethanol as pale yellow-green crystals (76%), m.p. 80–81°C (P^+ at *m/e* 251). Calculated for $C_{16}H_{13}NO_2$: C, 76·47; H, 5·22; N, 5·58. Found: C, 76·35; H, 5·18; N, 5·67.

3.8 Ethyl 2-cyano-3-(2-thienyl) acrylate (5g)

Compound **5g** was prepared in a manner similar to that described above for **5c**. It was recrystallized from ethanol as pale orange crystals (76%), m.p. 89–93°C (P^+ at *m/e* 207). Calculated for $C_{10}H_9NO_2S$: C, 57·96; H, 4·38; N, 6·76. Found: C, 57·84; H, 4·29; N, 6·55.

3.9 Ethyl 2-cyano-3-[4-(2-cyanostyryl)benzyl] acrylate (5h)

Compound **5h** was prepared in a manner similar to that described above for **5c**. It was recrystallized from ethanol as yellow-green crystals (80%), m.p. 169–172°C (P^+ at *m/e* 328). Calculated for $C_{21}H_{16}N_2O_2$: C, 76·81; H, 4·91; N, 8·53. Found: C, 76·80; H, 4·86; N, 8·44.

3.10 3-Cyano-4-phenyl-6-pyrenyl-2-pyridone (7a)

A mixture of ethyl 2-cyano-3-phenyl acrylate (**5a**; 2·01 g, 0·01 mole), 3-acetylpyrene (**6**; 2·44 g, 0·01 mole) and ammonium acetate (2·5 g) in ethanol (15 ml) was heated at reflux. During the reaction, the yellow

product precipitated. The crude product was filtered, washed with acetone and recrystallized from chloroform as yellow crystals (36%), m.p. 290–294°C (P^+ at *m/e* 396). Calculated for $C_{28}H_{16}N_2O$: C, 84·83; H, 4·07; N, 7·07. Found: C, 83·94; H, 4·07; N, 6·90.

3.11 3-Cyano-4-(4-methoxyphenyl)-6-pyrenyl-2-pyridone (7b)

Compound **7b** was prepared in a manner similar to that described above for **7a**. It was recrystallized from chloroform as yellow crystals (45%), m.p. 315–320°C (P^+ at *m/e* 426). Calculated for $C_{29}H_{18}N_2O_2$: C, 81·67; H, 4·25; N, 6·57. Found: C, 80·84; H, 4·28; N, 6·25.

3.12 3-Cyano-4-(4-methylphenyl)-6-pyrenyl-2-pyridone (7c)

Compound **7c** was prepared in a manner similar to that described above for **7a**. It was recrystallized from chloroform as yellow crystals (36%), m.p. 317–318°C (P^+ at *m/e* 410). Calculated for $C_{29}H_{18}N_2O$: C, 84·86; H, 4·42; N, 6·82. Found: C, 84·73; H, 4·44; N, 6·35.

3.13 3-Cyano-4-(4-cyanophenyl)-6-pyrenyl-2-pyridone (7d)

Compound **7d** was prepared in a manner similar to that described above for **7a**. It was recrystallized from chloroform as orange crystals (50%), m.p. 274–278°C (P^+ at *m/e* 421). Calculated for $C_{29}H_{15}N_3O$: C, 82·65; H, 3·59; N, 9·97. Found: C, 81·84; H, 3·55; N, 9·45.

3.14 3-Cyano-4-styryl-6-pyrenyl-2-pyridone (7e)

Compound **7e** was prepared in a manner similar to that described above for **7a**. It was recrystallized from chloroform as yellow crystals (26%), m.p. 333–335°C (P^+ at *m/e* 422). Calculated for $C_{30}H_{18}N_2O$: C, 85·29; H, 4·29; N, 6·63. Found: C, 84·97; H, 4·29; N, 6·25.

3.15 3-Cyano-4-(1-naphthyl)-6-pyrenyl-2-pyridone (7f)

Compound **7f** was prepared in a manner similar to that described above for **7a**. It was recrystallized from chloroform as yellow-green crystals (25%), m.p. 305–309°C (P^+ at *m/e* 446). Calculated for $C_{32}H_{18}N_2O$: C, 86·08; H, 4·06; N, 6·27. Found: C, 86·00; H, 4·12; N, 6·09.

3.16 3-Cyano-4-(2-thienyl)-6-pyrenyl-2-pyridone (7g)

Compound **7g** was prepared in a manner similar to that described above for **7h**. It was recrystallized from chloroform as yellow crystals (55%),

m.p. >350°C (P^+ at *m/e* 402). Calculated for $C_{26}H_{14}N_2OS$: C, 77·59; H, 3·51; N, 6·96. Found: C, 77·10; H, 3·61; N, 6·85.

3.17 3-Cyano-4-[4-(2-cyanostyryl)benzyl]-6-pyrenyl-2-pyridone (7h)

Compound **7h** was prepared in a manner similar to that described above for **7a**. It was recrystallized from chloroform as yellow crystals (35%), m.p. 325–330°C (P^+ at *m/e* 523). Calculated for $C_{37}H_{21}N_3O$: C, 84·88; H, 4·04; N, 8·03. Found: C, 84·43; H, 4·26; N, 8·01.

3.18 2-Chloro-3-cyano-4-phenyl-6-pyrenylpyridine (8a)

A mixture of 3-cyano-4-phenyl-6-pyrenyl-2-pyridone (**7a**; 1 g, 0·0025 mole), $POCl_3$ (30 g) and PCl_5 (0·6 g) was heated at reflux for 20 h, after which the mixture was stirred into ice water. The resulting precipitate was filtered, washed neutral and recrystallized from DMSO to give yellow-green crystals (90%), m.p. 231–235°C (P^+ at *m/e* 414·5). Calculated for $C_{28}H_{15}N_2Cl$: C, 81·06; H, 3·62; N, 6·76. Found: C, 80·96; H, 3·70; N, 6·47.

3.19 2-Chloro-3-cyano-4-(4-methoxyphenyl)-6-pyrenylpyridine (8b)

Compound **8b** was prepared in a manner similar to that described above for **8a**. It was recrystallized from DMSO as yellow-green crystals (87%), m.p. 220–224°C (P^+ at *m/e* 444·5). Calculated for $C_{29}H_{17}N_2OCl$: C, 78·29; H, 3·82; N, 6·30. Found: C, 78·01; H, 3·91; N, 6·20.

3.20 1-Methyl-3-cyano-4-phenyl-6-pyrenyl-2-pyridone (9a)

A mixture of 3-cyano-4-phenyl-6-pyrenyl-2-pyridone (**7a**; 0·5 g, 0·013 mole) in ethanol (40 ml), NaOH (0·4 g) in H_2O (1 ml) and methyl iodide (3 ml) was refluxed for 20 h, after which the solvent was vacuum evaporated and the residue washed with water to give a yellow product. Recrystallisation from DMSO gave yellow-green crystals (70%), m.p. 198–199°C (P^+ at *m/e* 410). Calculated for $C_{29}H_{18}N_2O$: C, 84·88; H, 4·39; N, 6·83. Found: C, 84·38; H, 4·84; N, 6·58.

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