

The brown precipitate was washed with ¹PrOH/hexane 1 : 4 and dried *in vacuo* to give **10**·Cl[−] (136 mg, 89%) as a reddish brown powder. M.p. >264° (dec.). ¹H-NMR (250 MHz, CD₃OD): 8.03 (*dd*, ³*J*=4.8, ⁴*J*=1.5, 2 H); 8.13 (*dd*, ³*J*=4.6, ⁴*J*=1.5, 2 H); 8.79 (*d*, ³*J*=6.9, 2 H); 8.92 (*dd*, ³*J*=4.7, ⁴*J*=1.5, 2 H); 9.03 (*dd*, ³*J*=4.7, ⁴*J*=1.5, 2 H); 9.50 (*d*, ³*J*=6.9, 2 H). API-ES-MS (*pos.*): 234.2 (*M*⁺), 235.3.

1-Benzyl-1'-(4-[[1-benzopyridinium-4-yl]methyl]phenyl)-4,4'-bipyridinium Tris(hexafluorophosphate) (11·3 PF₆[−]). Benzyl bromide (5 mmol, 0.855 g) was added to a soln. of **9**·Cl[−] (0.5 mmol, 0.180 g) in ¹PrOH (5 ml) and refluxed for 18 h to yield a yellow precipitate. The latter was isolated, dissolved in H₂O and precipitated with 10% aq. NH₄PF₆ soln. The white product was isolated and dried *in vacuo*: 0.341 g (72%) of **11**·3 PF₆[−]. White powder. M.p. 221°. ¹H-NMR (250 MHz, CD₃OD): 4.58 (*s*, 2 H); 5.83 (*s*, 2 H); 6.03 (*s*, 2 H); 7.67–7.50 (*m*, 10 H); 7.78 (*d*, ³*J*=8.8, 2 H); 7.94 (*d*, ³*J*=8.5, 2 H); 8.05 (*d*, ³*J*=7.0, 2 H); 8.74 (*d*, ³*J*=7.0, 2 H); 8.80 (*d*, ³*J*=7.0, 2 H); 8.98 (*d*, ³*J*=7.0, 2 H); 9.38 (*d*, ³*J*=7.3, 2 H); 9.48 (*d*, ³*J*=7.0, 2 H).

1-Benzyl-1'-(1-benzylpyridinium-4-yl)-4,4'-bipyridinium Tris(hexafluorophosphate) (12·3 PF₆[−]). Benzyl bromide (0.2 mmol, 342 mg) and **10**·Cl[−] (0.2 mmol, 54 mg) were refluxed in ¹PrOH (3 ml) for 20 h. The precipitate was isolated, dissolved in H₂O and filtered to eliminate a brown by-product. The product was then precipitated with 10% aq. NH₄PF₆ soln. (3 ml), isolated, and dried *in vacuo*: 56.5 mg (33%) of **12**·3 PF₆[−]. Beige powder. M.p. >205° (dec.).

Synthesis of 14–20. General Procedure C. The monoaryl compound (0.01 mol) was treated with tosylate **13** (0.015 mol) in MeCN and stirred under reflux for 48 h. The solvent was evaporated and the residue dissolved in a few ml of H₂O, washed with Et₂O (4×50 ml), and evaporated. The solids were dissolved in MeOH, and the soln. was added dropwise to 3M aq. NH₄PF₆.

1-(2,4-Dinitrophenyl)-1'-(4-hydroxyphenyl)-4,4'-bipyridinium Bis(hexafluorophosphate) (14·2 PF₆[−]): Yield 26%. Yellowish powder. M.p. 212°. ¹H-NMR (250 MHz, CD₃CN): 7.08 (*d*, ³*J*=8.8, 2 arom. H); 7.57 (*d*, ³*J*=8.8, 2 arom. H); 8.09 (*d*, ³*J*=8.6, 1 arom. H); 8.54 (*d*, ³*J*=6.5, 2 H, Vio); 8.62 (*d*, ³*J*=6.7, 2 H, Vio); 8.79 (*d*, ³*J*=8.5, 1 arom. H); 9.08 (*d*, ³*J*=6.5, 4 H, Vio); 9.13 (*s*, 1 arom. H).

1-(2,4-Dinitrophenyl)-1'-(4-(hydroxymethyl)phenyl)-4,4'-bipyridinium Bis(hexafluorophosphate) (15·2 PF₆[−]): Yield 43%. Ochre. M.p. 241°. ¹H-NMR (250 MHz, D₂O): 7.65 (*d*, ³*J*=8.6, 2 arom. H); 7.73 (*d*, ³*J*=8.7, 2 arom. H); 8.19 (*d*, ³*J*=8.5, 1 arom. H); 8.71 (*d*, ³*J*=6.1, 2 H, Vio); 8.78 (*d*, ³*J*=3.6, 2 H, Vio); 8.84 (*d*, ³*J*=8.2, 1 arom. H); 9.33 (*d*, ³*J*=5.2, 5 H, arom. H, Vio).

1-(4-Carboxyphenyl)-1'-(2,4-dinitrophenyl)-4,4'-bipyridinium Bis(hexafluorophosphate) (16·2 PF₆[−]): Yield 58%. Beige powder. M.p. 222°. ¹H-NMR (250 MHz, D₂O): 7.8 (*d*, ³*J*=8.5, 2 arom. H); 8.05 (*d*, ³*J*=8.4, 2 arom. H); 8.19 (*d*, ³*J*=8.7, 1 arom. H); 8.85–8.71 (*m*, 5 H, arom. H, Vio); 9.38–9.31 (*m*, 5 H, arom. H, Vio).

1-(2,4-Dinitrophenyl)-1'-(4-sulfohenyl)-4,4'-bipyridinium Bis(hexafluorophosphate) (17·2 PF₆[−]): Yield 40.5%. Orange powder. M.p. >270° (dec.). ¹H-NMR (250 MHz, D₂O): 7.09 (*d*, ³*J*=8.6, 2 arom. H); 7.65 (*d*, ³*J*=8.1, 2 arom. H); 7.87 (*br.*, arom. H); 8.05 (*br.*, arom. H); 8.18 (*d*, arom. H); 8.57 (*d*, arom. H); 8.83 (*br.*, arom. H); 9.20 (*d*, arom. H); 9.30–9.38 (*br.*, arom. H).

1-(2,4-Dinitrophenyl)-1'-(4-ethylphenyl)-4,4'-bipyridinium Bis(hexafluorophosphate) (18·2 PF₆[−]): Yield 58.5%. White powder. M.p. 255°. ¹H-NMR (250 MHz, CD₃CN): 7.56 (*br.*, 2 arom. H); 7.63 (*br.*, 2 arom. H); 8.13 (*d*, ³*J*=7.0, 1 arom. H); 8.64 (*br.*, 4 H, Vio); 8.79 (*d*, ³*J*=7.07, 1 arom. H); 9.13 (*br.*, 5 H, arom. H, Vio). ¹³C-NMR (63 MHz, (D₆)DMSO): 16.3; 28.7; 122.4; 125.5; 127.5; 127.7; 130.4; 131.2; 132.8; 146.7; 148.1; 148.6; 149.0; 150.2.

1-(2,4-Dinitrophenyl)-1'-(4-ethoxyphenyl)-4,4'-bipyridinium Bis(hexafluorophosphate) (19·2 PF₆[−]): Yield 35%. Yellow. M.p. 254°. ¹H-NMR (250 MHz, CD₃CN): 1.36 (*t*, ³*J*=6.5, 3 H, Me); 4.12 (*q*, ³*J*=6.8, 2 H, CH₂); 7.17 (*d*, ³*J*=8.1, 2 arom. H); 7.64 (*d*, ³*J*=7.0, 2 arom. H); 8.08 (*d*, ³*J*=7.2, 1 arom. H); 8.58 (*d*, ³*J*=20.5, 4 H, Vio); 8.79 (*d*, ³*J*=7.1, 1 arom. H); 9.08 (*br.*, 5 H, Vio, arom. H).

1-[4-[(Diethoxyphosphinyl)methyl]phenyl]-1'-(2,4-dinitrophenyl)-4,4'-bipyridinium Bis(hexafluorophosphate) (20·2 PF₆[−]): Yield 25%. White powder. M.p. 209°. ¹H-NMR (250 MHz, CD₃CN): 1.30 (*t*, ³*J*=7.1, 2 Me); 3.40 (*d*, ³*J*=22.0, 1 CH₂); 4.08 (*q*, ³*J*=7.3, 2 CH₂); 7.77 (*m*, 4 arom. H); 8.20 (*d*, ³*J*=8.8, 1 arom. H); 8.73 (*m*, 4 H, Vio); 8.90 (*d*, ³*J*=8.4, 1 arom. H); 9.24–9.17 (*m*, 5 H, Vio, arom. H).

1-[4-[(Diethoxyphosphinyl)methyl]phenyl]-1'-(4-fluorophenyl)-4,4'-bipyridinium Bis(hexafluorophosphate) (21·2 PF₆[−]). Bis(hexafluorophosphate) **20** (5 mmol) was treated with 4-fluorobenzeneamine (7.5 mmol) in 80% EtOH under reflux for 24 h. The mixture was evaporated, the residue dissolved in H₂O, and the soln. washed with Et₂O (4×). After anion exchange with aq. 3M NH₄PF₆, the precipitate was filtered and dried *in vacuo*. ¹H-NMR (D₂O): 3.15 (*d*, 2 H); 7.1–7.7 (*m*, 8 H); 8.6–9.3 (*m*, 8 H).

1-[4-[(Diethoxyphosphinyl)methyl]phenyl]-1'-(4-methylphenyl)-4,4'-bipyridinium Bis(hexafluorophosphate) (22·2 PF₆[−]): As described for **21**, with 4-methylbenzeneamine. ¹H-NMR (D₂O): 2.33 (*s*, 3 H); 3.25 (*d*, 2 H); 7.35–7.8 (*m*, 8 H); 8.7–9.3 (*m*, 8 H).