Synthesis of 23–26: General Procedure D. The 1-aryl-substituted 1'-(2,4-dinitrophenyl)-4,4'-bipyridinium precursor (5 mmol) was treated with 4-aminobenzenmethanol (7.5 mmol) in 80% aq. EtOH under reflux for 24 h. The soln. was evaporated, and the residue dissolved in H_2O (50 ml), the soln. washed with Et_2O (4×30 ml) and evaporated, and the residue dissolved in MeOH (3 ml) and ion-exchanged with aq. $3M NH_4PF_6$.

1'-[4-(Hydroxymethyl)phenyl]-1-(4-hydroxyphenyl)-4,4'-bipyridinium Bis(hexafluorophosphate) (23 · 2 PF $_6$): Yield 57%. Reddish-brown powder. M.p. >265° (dec.). ¹H-NMR (250 MHz, CD $_3$ CN): 4.80 (s, 2 H, CH $_2$); 7.19 (d, 3J =8.82, 2 arom. H); 7.67 (d, 3J =8.57, 2 arom. H); 7.78 (s, 4 arom. H); 8.65 (t, 3J =6.95, 4 H, Vio); 9.16 (d, 3J =6.7, 2 H, Vio); 9.22 (d, 3J =6.82, 2 H, Vio).

1-(4-Ethylphenyl)-1'-[4-(hydroxymethyl)phenyl]-4,4'-bipyridinium Bis(hexafluorophosphate) ($24 \cdot 2 \text{ PF}_6$): Yield 53%. Pale yellow powder. M.p. 230°. ¹H-NMR (250 MHz, CD₃CN): 1.23 (t, ³J=7.5, Me); 2.76 (q, ³J=7.5, 1 CH₂); 4.68 (s, 1 CH₂); 7.54 (d, ³J=8.4, 2 arom. H); 7.66–7.61 (m, 6 arom. H); 8.55 (d, ³J=5.5, 4 H, Vio); 9.10 (d, ³J=4.1, 4 H, Vio).

1-(4-Ethoxyphenyl)-1'-[4-(hydroxymethyl)phenyl]-4,4'-bipyridinium Bis(hexafluorophosphate) (25 · 2 PF₆): Yield 84%. Yellow powder. M.p. 265°. ¹H-NMR (250 MHz, CD₃CN): 1.35 (t, ${}^{3}J$ =7.3, Me); 4.11 (q, ${}^{3}J$ =7.1, 1 CH₂); 4.65 (s, 1 CH₂); 7.16 (d, ${}^{3}J$ =9.4, 2 arom. H); 7.63 (d, ${}^{3}J$ =9.3, 2 arom. H); 7.76−7.66 (m, 4 arom. H); 8.54 (m, 4 H, Vio); 9.11−9.05 (m, 4 H, Vio).

1-(4-Carboxyphenyl)-1'-[4-(hydroxymethyl)phenyl]-4,4'-bipyridinium Bis(hexafluorophosphate) (26- 2 PF_6^-): Yield 65.5%. Dark yellow powder. M.p. $> 270^\circ$ (dec.). ¹H-NMR (250 MHz, CD₃CN): 4.64 (*d*, ${}^3J=27.1$, 1 CH₂); 7.82 (*d*, ${}^3J=8.5$, 2 arom. H); 8.29 (*d*, ${}^3J=8.5$, 2 arom. H); 8.59 (*d*, ${}^3J=6.3$, 4 H, Vio); 9.13 (*m*, 4 H, Vio).

Synthesis of the Phosphonic Acids 28–34: General Procedure E. 1-[2-(Diethoxyphosphinyl)ethyl]-1'-(2,4-dinitrophenyl)-4,4'-bipyridinium (27; 0.005 mol) prepared according to [13], was added to the appropriately substituted aromatic amine (0.075 mol) in EtOH (60 ml). The solvent was evaporated, and H₂O (80 ml) was added. The suspension was stirred and filtered, and the filtrate decolorized with charcoal and then evaporated. The resulting product was dissolved in MeCN and the solid filtered and dried *in vacuo* to yield the phosphonate ester derivative. The latter was refluxed in 50% HCl soln. (60 ml) for 24 h. Then, the solvent was evaporated and the residue dried *in vacuo*: phosphonic acid derivative. For spectroscopic data for 28–34, see also [22].

 $\begin{array}{ll} \textit{1-(2-Phosphonoethyl)-1'-[4-(pyridine-4-ylmethyl)phenyl]-4,4'-bipyridinium} & \textit{Hexafluorophosphate} \\ \textbf{(28\cdot PF}_6^-): Yield 95\%. \ White powder. \ M.p. 211°. \ ^1H-NMR (250 \ MHz,D_2O): 2.32 \ (br., 1 \ CH_2); 4.35 \ (s, 1 \ CH_2); 7.57 \ (d, 2 \ arom. \ H); 7.71 \ (m, 4 \ arom. \ H); 8.59 \ (d, 2 \ H, \ Vio); 9.08 \ (d, 2 \ H, \ Vio); 9.25 \ (d, 2 \ H, \ Vio). \end{array}$

1-(4-Cyanophenyl)-1'-(2-phosphonoethyl)-4,4'-bipyridinium Dichloride (29 · 2 Cl $^{-}$): ¹H-NMR (D₂O): 2.3 (m, 2 H); 4.8 (m, 2 H); 7.8 (d, 2 H); 8.16 (d, 2 H); 8.4−9.2 (m, 8 H).

1-[4-(tert-Butyl)phenyl]-1'-(2-phosphonoethyl)-4,4'-bipyridinium Dichloride (30·2 Cl[−]): ¹H-NMR (D₂O): 1.29 (s, 9 H); 2.31 (m, 2 H); 4.78 (m, 2 H); 7.60 (d, 2 H); 7.68 (d, 2 H); 8.56−9.2 (m, 8 H).

1-(4-Methylphenyl)-1'-(2-phosphonoethyl)-4,4'-bipyridinium Dichloride ($31 \cdot 2 \text{ Cl}^-$): $^1\text{H-NMR}$ (D₂O): 2.32 (s, 3 H); 2.4–2.52 (m, 2 H); 4.75 (m, 2 H); 7.42 (d, 2 H); 7.53 (d, 2 H); 8.48–9.19 (m, 8 H).

1-(4-Phenoxyphenyl)-1'-(2-phosphonoethyl)-4,4'-bipyridinium Dichloride (32 · 2 Cl[−]): 1 H-NMR (CD₃CN, PF₆): 2.36 (m, 2 H); 4.88 (m, 2 H); 7.1–7.45 (m, 5 H); 7.44–7.74 (m, 4 H); 8.49–9.11 (m, 8 H).

1-(4-Fluorophenyl)-I⁻(2-phosphonoethyl)-4,4'-bipyridinium Dichloride (33·2 Cl⁻): ¹H-NMR (D₂O): 2.33 (m, 2 H); 4.81 (m, 2 H); 7.35 (d, 2 H); 7.71 (d, 2 H); 8.58–9.22 (m, 8 H).

1-(4-Benzoylphenyl)-1'-(2-phosphonoethyl)-4,4'-bipyridinium Dichloride ($34 \cdot 2 \text{ Cl}^-$): ${}^1\text{H-NMR}$ ($D_2\text{O}$): 2.32 (m, 2 H); 4.8 (m, 2 H); 7.4-8.2 (m, 9 H); 8.40-9.33 (m, 8 H).

REFERENCES

- [1] T. V. Laurinavichene, N. A. Zorin, A. A. Tsygankov, Arch. Microbiol. 2002, 178, 437.
- [2] I. Ichinose, T. Kunitake, Adv. Mater. 2002, 14, 344.
- [3] S. Heinen, W. Meyer, L. Walder, J. Electroanal. Chem. 2001, 498, 34.
- [4] Y. Chen, G. Y. Jung, D. A. A. Ohlberg, X. M. Li, D. R. Stewart, J. O. Jeppesen, K. A. Nielsen, J. F. Stoddart, R. S. Williams, *Nanotechnology* 2003, 14, 462.
- [5] G. D. Sharma, D. Saxena, M. S. Roy, Synth. Met. 1999, 106, 97.
- [6] F. Campus, P. Bonhote, M. Gratzel, S. Heinen, L. Walder, Sol. Energy Mater. Sol. Cells 1999, 56, 281.
- [7] M. T. Moller, S. Asaftei, D. Corr, M. Ryan, L. Walder, Adv. Mater. 2004, 16, 1558.
- [8] F. N Castellano, G. J. Meyer, Mol. Lev. Artif. Photosynth. Mat. 1997, 44, 167.
- [9] A. Yasuda, H. Mori, J. Mizuguchi, Jpn. J. Appl. Phys., Part 1, Regul. Pap. Short Notes Rev. Pap. 1987, 26, 1352.