

P. Molina*, M. Alajarin, M. Lorenzo-Peña, A. Tárraga and M. J. Vilaplana

Departamento de Química Orgánica, Facultad de Ciencias, Universidad de Murcia,

Murcia, Spain

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β -Functionalized primary amines are converted by the 2-ethoxycarbonyl-4,6-diphenylpyrylium cation into the corresponding bicyclic salts **2**. The *N*-substituted 4,6-diphenylpyridine-2-thiones **6** undergo cyclization to the bicyclic compounds **8** by action of methyl iodide.

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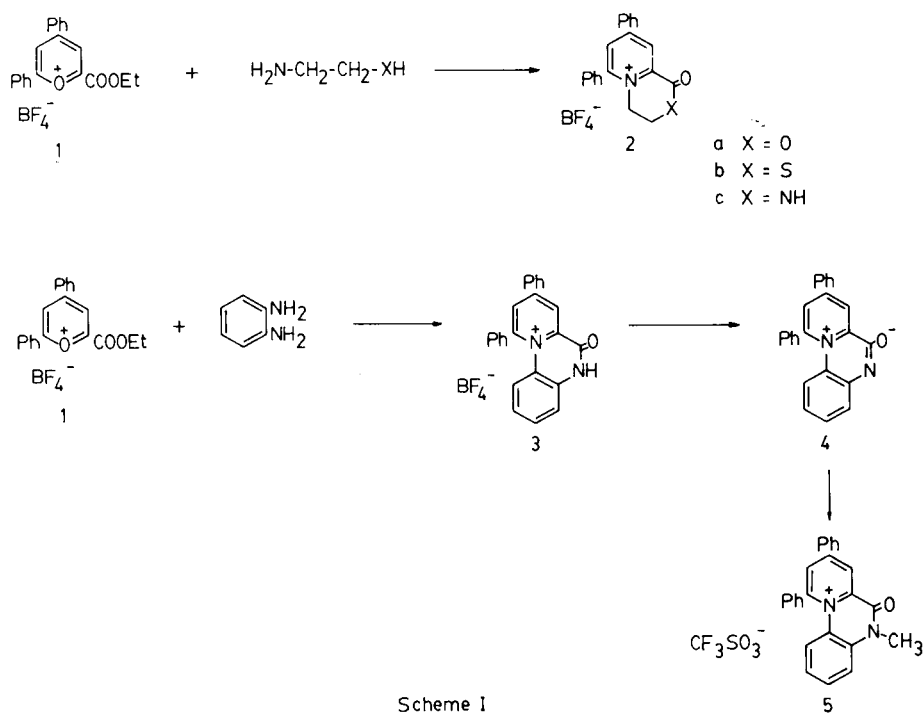
The reaction of several pyrylium salts with β -functionalized amines such as 2-chloroethylamine, ethanolamine and *N,N*-diethylethylenediamine to give the corresponding pyridinium salts have been described [1,2].

In this context, we have previously reported the reaction of 2-functionalized pyrylium salts with several nitrogen nucleophiles to give bridgehead nitrogen heterocycles. 2-Ethoxycarbonyl-4,6-diphenylpyrylium cation reacts with *N*³-aryl substituted and unsubstituted amidrazones to give 2,3-diaryl-6,8-diphenyl-4-oxopyrido[2,1-*f*]-1,2,4-triazinium salts and 2-substituted 4-oxido-6,8-diphenylpyrido[2,1-*f*]-1,2,4-triaziniums respectively [3,4], while with aldehyde and ketone hydrazones [5] or thiocarbonylhydrazide [6] yielded 7,7-disubstituted 2,3a-diphenyl-3,3a-dihydro-7*H*-pyrazolo[1,5-*c*]-1,3-oxazines and 3-amino-6,8-diphenyl-4-oxopyrido[2,1-*f*]-1,2,4-triazin-9-ium-2-thioxide respectively.

On the other hand, 2-methylthio-4,6-diphenylpyrylium cation reacts with *N*³-arylamidrazones to give the cor-

responding 1,2-diaryl-5,7-diphenyl-1,2,4-triazolo[1,5-*a*]pyridinium salts [3,4].

In connection with our investigation on the preparation of pharmacologically interesting fused heterocycles such as 1,2,4-triazolo[1,5-*a*]pyridine [7], pyrazolo[1,5-*a*]pyridine [8,9,10] or 1,3,4-oxadiazolo[3,2-*a*]pyridine derivatives and thioanalogues [11,12] we report here attempts to synthesize bridgehead nitrogen heterocycles such as 1-oxo-1,2,3,4-tetrahydropyrido[1,2-*a*]pyrazin-5-ium, 1-oxo-3,4-dihydro-1*H*-pyrido[2,1-*c*]-1,4-oxazin-5-ium, 1-oxo-3,4-dihydro-1*H*-pyrido[2,1-*c*]-1,4-thiazin-5-ium and 6-oxido-8,10-diphenylpyrido[1,2-*a*]quinoxalin-1-ium by reaction of 2-ethoxycarbonyl-4,6-diphenylpyrylium tetrafluoroborate with β -functionalized amines. Another approach is based in the cyclization of *N*-substituted 2-methylthio-4,6-diphenylpyridinium cations to 2,3-dihydrooxazolo[3,2-*a*]pyridine, 2,3-dihydrothiadiazolo[3,2-*a*]pyridine and 2,3-dihydro-1*H*-imidazolo[3,2-*a*]pyridine derivatives. Some of these hetero-



Scheme 1

cyclic ring systems possess a variety of pharmacological activities *e.g.* derivatives of the pyrido[1,2-*a*]pyridazine ring system show neurotropic [13], anticholinergic [14] and antiarrhythmic [15] activities; derivatives of the thiazolo[3,2-*a*]pyridine ring system possess pharmacological activities like antiinflammatory [16], hypoglycemic [17] and analgesic [18] activity.

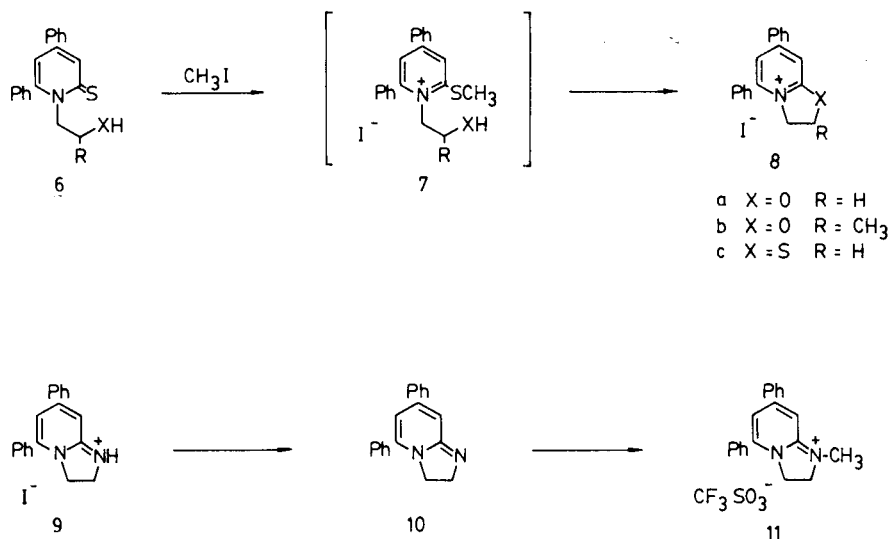
The pyrylium salt **1** reacts with β -functionalized amines such as ethanolamine, cysteamine and ethylenediamine at room temperature in ethanol in the presence of triethylamine giving the corresponding bicyclic salts **2** as crystalline solids in moderate to good yields (43-83%) (Table I). The ir spectra of these bicyclic salts show an absorption at 1650-1750 cm^{-1} which can be attributed to the C=O stretching vibration. Proton nmr spectral data for the bicyclic salts **2** are collected in Table II. In general, the characteristic finely splitted doublet pattern is shown for the 3,5-protons of the pyridinium ring; the other aryl signals give the expected multiplets and the characteristic pattern is found for the $\text{N}^+\text{-CH}_2\text{-CH}_2\text{-X}$ group. In particular, for **2a** these protons appear as a narrow multiplet at δ 4.95, for **2b** as two triplets centered at δ 3.65 and 5.00 respectively, and for **2c** as two triplets centered at δ 3.95 and 4.75 respectively. Mass spectra of compounds **2** are in accordance with the proposed structures.

Compound **1** reacts with *o*-phenylenediamine at room temperature in methanolic solution to give the salt **3**, whereas in the presence of triethylamine leads to the zwitterionic compound **4**, which undergoes *N*-methylation by the action of methyl trifluoromethanesulfonate to give **5**. The ir spectra of compounds **3** and **5** show an absorption at 1710 cm^{-1} attributed to the C=O stretching vibration,

which is absent in **4**. The proton nmr spectrum of **5** shows among others a signal as singlet at δ 3.30 due to the N- CH_3 group. Mass spectra of compounds **3** and **5** show the fragment ($\text{M}^+ \text{-HX}$) whereas compound **4** shows the expected molecular ion peak.

Pyridine-2-thiones **6**, readily available from 4,6-diphenyl-2*H*-pyran-2-thione and β -functionalized amines such as ethanolamine, 2-hydroxypropylamine and cysteamine, react with methyl iodide in dry benzene at room temperature to give the bicyclic salts **8** as crystalline solids in moderate to good yields (Table I). We believe that the mechanism involves the formation of the *N*-substituted 4,6-diphenyl-2-methylthiopyridinium cation **7** as intermediate, which undergoes intramolecular nucleophilic displacement to give the corresponding pyridinium salts **8** and methanethiol. The proton nmr spectra of compounds **8** are consistent with their formulation as bicyclic salts. Thus, the signals corresponding to the methylene groups appear as a multiplet centered at δ 5.2 for **8a**, while for **8c** appear as two triplets at δ 4.95 and 3.72 respectively. In **8b** the methine proton appears as a multiplet centered at δ 6.0, and the methylene protons as two multiplets at δ 5.45 and 4.55 respectively. The mass spectra of **8a** and **8b** show the expected molecular ion and the fragmentation pattern are in accordance with the proposed structures. In compound **8c** the fragment at m/e 290 ($\text{M}^+ \text{-HBF}_4$) is the base peak.

Similarly, pyridine 2-thione **6** ($\text{R} = \text{H}$, $\text{X} = \text{NH}$) by action of methyl iodide undergoes cyclization to the salt **9**, which in basic medium is converted into the neutral biheterocycle **10**. This compound undergoes methylation by reaction with methyl trifluoromethanesulfonate to give **11**.



Scheme II

Table I
Bridgehead Nitrogen Heterocycles Prepared

| No. | Yield (%) | Mp (°C) | Appearance Solvent | Molecular Formula | Analyses (%) | | | Found | | |
|-----------|-----------|---------|--------------------------------------|--|--------------|------|-------|-------|------|-------|
| | | | | | C | H | N | C | H | N |
| 2a | 50 | 196-198 | white needles methanol | C ₂₀ H ₁₆ BF ₄ NO ₂ (388.95) | 61.75 | 4.11 | 3.59 | 61.87 | 4.00 | 3.64 |
| 2b | 43 | 246-247 | yellow prisms ethanol-ether | C ₂₀ H ₁₆ BF ₄ NOS (405.02) | 59.30 | 3.95 | 3.45 | 59.45 | 3.80 | 3.57 |
| 2c | 83 | 279-280 | white-prisms methanol | C ₂₀ H ₁₇ BF ₄ N ₂ O (387.96) | 61.91 | 4.38 | 7.21 | 61.79 | 4.31 | 7.30 |
| 3 | 70 | 303-305 | yellow prisms ethanol | C ₂₄ H ₁₇ BF ₄ N ₂ O (436.0) | 66.11 | 3.89 | 6.42 | 66.30 | 3.78 | 6.40 |
| 4 | 68 | 155-157 | yellow needles ethanol-chloroform | C ₂₄ H ₁₆ N ₂ O (348.23) | 82.77 | 4.59 | 8.04 | 82.69 | 4.50 | 7.95 |
| 5 | 72 | 168-171 | white prisms dichloromethane | C ₂₆ H ₁₉ F ₃ N ₂ O ₄ S (512.25) | 60.95 | 3.70 | 5.46 | 60.85 | 3.59 | 5.40 |
| 8a | 79 | 213-215 | yellow-prisms methanol-ether | C ₁₉ H ₁₆ INO (401.25) | 56.87 | 4.02 | 3.49 | 56.75 | 3.99 | 3.56 |
| 8b | 46 | 253-255 | yellow prisms methanol-ether | C ₂₀ H ₁₆ INO (415.27) | 57.84 | 4.37 | 3.37 | 57.81 | 4.29 | 3.32 |
| 8c | 45 | 226-228 | yellow prisms methanol-ether | C ₁₉ H ₁₆ BF ₄ NS (377.21) | 60.50 | 4.27 | 3.71 | 60.58 | 4.30 | 3.62 |
| 9 | 48 | 133-135 | yellow prisms ethanol-ether | C ₁₉ H ₁₇ IN ₂ (400.25) | 57.01 | 4.28 | 6.99 | 56.87 | 4.33 | 7.08 |
| 10 | 81 | 148-150 | orange prisms ethanol-ether | C ₁₉ H ₁₆ N ₂ (272.34) | 83.79 | 5.92 | 10.28 | 83.64 | 6.08 | 10.16 |
| 11 | 93 | 150-152 | yellow needles ethanol | C ₂₁ H ₁₉ F ₃ N ₂ O ₃ S (436.44) | 57.79 | 4.39 | 6.41 | 57.86 | 4.26 | 6.44 |

The proton nmr spectra of **9** and **11** do not show signals of pyridinium ring protons. However the signals corresponding to the 3,5-protons of the pyridine ring appear as two doublets ($J = 2$ Hz) near to the aromatic region; the characteristic pattern is found for the N-CH₂-CH₂-N group. In particular, for **10** the methylene protons appear as a narrow multiplet at δ 4.05, while for **9** and **11** appear as two multiplets; in addition, compound **11** shows a singlet at δ 3.20 due to the N-CH₃ group. Mass spectra of compounds **9** and **11** show the expected fragment ($M^+ -HX$), while compound **10** shows the molecular ion peak (Table II).

EXPERIMENTAL

The melting points were determined with a Kofler hot stage microscope and were uncorrected. The ir spectra were recorded of mineral oil mulls with a Nicolet-FT 5DX instrument. The ¹H-nmr spectra were recorded with a Varian EM-360 instrument with TMS as internal standard. Mass spectra were obtained with a Hewlett-Packard 5993 C GC/MS system; compounds were introduced through the direct insertion probe. The electron beam energy was 70 eV and the ion source was at ca 300°. Microanalyses were performed with a Perkin-Elmer 240 C instrument.

Reagents.

2-Ethoxycarbonyl-4,6-diphenylpyrylium tetrafluoroborate (**1**) was prepared from ethyl pyruvate, benzylideneacetophenone and boron trifluoride-diethyl ether [19]. 4,6-Diphenylpyran-2-thione was prepared from 4,6-diphenyl-2-pyrone and phosphorus pentasulfide [20].

Bicyclic Salts **2**. General Procedure.

2-Ethoxycarbonyl-4,6-diphenylpyrylium tetrafluoroborate (**1**) (10 mmoles) was dissolved in ethanol (150 ml), the appropriate β -functionalized amine (10 mmoles) was added. The reaction mixture was stirred at room temperature until a solid separated (15-17 hours), which when recrystallized from the appropriate solvent gave **2** (Table I).

6-Oxide-8,10-diphenylpyrido[1,2-*a*]quinoxalin-1-ium (**4**).

2-Ethoxycarbonyl-4,6-diphenylpyrylium tetrafluoroborate (**1**) (10 mmoles) was dissolved in methanol (150 ml), equimolecular amounts of *o*-phenylenediamine and triethylamine were added. The reaction mixture was stirred at room temperature for 6 hours and the yellow precipitated solid was separated by filtration and recrystallized from methanol-chloroform (1:4) to give **4** as yellow needles (Table I).

8,10-Diphenyl-5*H*-6-oxopyrido[1,2-*a*]quinoxalin-1-ium Tetrafluoroborate (**3**).

2-Ethoxycarbonyl-4,6-diphenylpyrylium tetrafluoroborate (**1**) (10 mmoles) was dissolved in methanol (150 ml), *o*-phenylenediamine (10 mmoles) was added and the reaction mixture was stirred at room temperature for 2 hours. The yellow precipitated solid was collected by filtration and recrystallized from methanol-chloroform to give **3** in 13% yield. From the mother liquor, the solvent was removed under reduced pressure and the residual material was treated with ether and the yellow solid formed was recrystallized from ethanol to give **3** as yellow prisms (Table I).

When compound **3** was treated with equimolecular amount of triethylamine in methanol at room temperature gave **4** in quantitative yield.

8,10-Diphenyl-5-methyl-6-oxopyrido[1,2-*a*]quinoxalin-1-ium Trifluoromethanesulphonate (**5**).

Table II
Spectral Data of Compounds **2-5** and **8-11**

| Compound No. | IR (cm ⁻¹) | ¹ H-NMR (δ ppm) [a] | MS m/e (%) |
|--------------|---|--|---|
| 2a | 3080, 1750, 1625, 1570, 1400 1300, 1220, 1170, 1050, 915, 785, 775, 770, 700, 690 | 9.10 (1H, d, J = 2 Hz), 8.85 (1H, d, J = 2 Hz), 7.6-8.45 (10H, m), 4.85 (4H, s) | 230 (50), 115 (21) 77 (20), 44 (100) |
| 2b | 1650, 1620, 1440, 1345, 1290 1060, 1000, 920, 910, 780, 775, 705, 695 | 8.80 (1H, d, J = 2 Hz), 8.40 (1H, d, J = 2 Hz), 7.5-8.1 (10H, m), 5.00 (2H, t) 3.65 (2H, t) | 317 (M ⁺ - HBF ₄ , 3), 285 (8), 230 (99), 115 (16), 77 (28), 60 (100), 32 (56) |
| 2c | 3360, 1700, 1620, 1560, 1175, 1060, 1000, 945, 910, 780, 770, 700, 690 | 9.60 (1H, s), 9.01 (1H, d, J = 2 Hz), 8.75 (1H, d, J = 2 Hz), 7.7-8.45 (10H, m) 4.75 (2H, t), 3.95 (2H, t) | 300 (M ⁺ - HBF ₄ , 70), 299 (100), 230 (84), 203 (39), 115 (18), 77 (37), 28 (15) |
| 3 | 1710, 1620, 1590, 1490, 1060 895, 770, 750, 730, 710 | 9.40 (1H, s), 8.90 (1H, d, J = 2 Hz), 8.70 (1H, d, J = 2 Hz), 7.2-8.3 (14H, m) | 348 (M ⁺ - HBF ₄ , 12), 320 (42), 273 (10), 230 (8), 115 (14), 105 (5), 77 (23) |
| 4 | 1675, 1620, 1590, 1530, 1450 1220, 1160, 900, 760, 690 655 | 9.10 (1H, d, J = 2 Hz), 8.60 (1H, d, J = 2 Hz), 7.2-8.3 (14H, m) | 348 (M ⁺ , 20), 320 (38) 273 (12), 230 (30), 115 (16), 105 (8), 77 (34) |
| 5 | 1710, 1620, 1540, 1495, 1295 1250, 1155, 1030, 900, 790, 780, 760 | 8.80 (1H, d, J = 2 Hz), 8.6 (1H, d, J = 2 Hz), 7.3-8.4 (14H, m), 3.30 (3H, s) | 363 (M ⁺ - CF ₃ SO ₃ , 5), 349 (34), 320 (100), 272 (11) 105 (5), 77 (10) |
| 8a | 3060, 1630, 1570, 1470, 1370, 1305, 1260, 1220, 1130, 1000, 970, 870, 770, 710, 690 | 7.4-8.0 (12H, m), 5.20 (4H, m, A ₂ B ₂ system) | 401 (M ⁺ , 36), 400 (20), 274 (70), 272 (11), 248 (19), 247 (100), 230 (11) 219 (25), 217 (11), 202 (10), 115 (18), 77 (10) |
| 8b | 1640, 1575, 1460, 1225, 1135 1040, 955, 890, 875, 860, 780, 690 | 7.4-8.2 (12H, m), 5.8-6.3 (1H, m) 5.2-5.7 (1H, m), 4.3-4.8 (1H, m) 1.70 (3H, d) | 415 (M ⁺ , 3), 288 (15), 287 (18), 286 (34), 272 (25), 254 (10), 248 (20), 247 (100), 219 (37), 191 (12), 127 (30), 115 (66), 102 (16), 89 (24), 77 (32) |
| 8c | 3060, 1615, 1550, 1500, 1430, 1290, 1260, 1225, 1180, 1160, 1060, 880, 840, 770, 735, 710 | 7.3-8.0 (12H, m), 4.95 (2H, t, $\overset{\dagger}{N}$ -CH ₂) 3.72 (2H, t, S-CH ₂) | 290 (M ⁺ - HBF ₄ , 100), 289 (33), 288 (72), 263 (71), 230 (46), 219 (23), 203 (53), 202 (62), 145 (29), 115 (87), 102 (38), 77 (82) |
| 9 | 3150, 1640, 1575, 1405, 1305 1030, 855, 770, 700, 690 | 9.45 (1H, s), 7.6-8.3 (10H, m) 7.50 (1H, d), 7.40 (1H, d), 4.5-5.0 (2H, m), 3.9-4.4 (2H, m) | 273 (M ⁺ - HI, 12), 272 (61) 271 (100), 230 (8), 215 (6), 202 (17), 128 (98), 127 (53), 115 (9), 77 (15) |
| 10 | 1640, 1550, 1495, 1275, 1175, 1160, 1100, 1075, 1030, 1010, 840, 770, 690 | 7.6-7.9 (10H, m), 6.90 (1H, d), 6.15 (1H, d), 4.05 (narrow m) | 272 (M ⁺ , 68), 271 (100) 244 (10), 230 (8), 203 (10), 202 (17), 135 (11), 128 (12), 115 (12), 77 (18) |
| 11 | 1650, 1580, 1570, 1465, 1455, 1270, 1160, 1145, 1030, 840, 765, 710, 680, 635 | 7.4-8.0 (10H, m), 7.0 (1H, d) 6.9 (1H, d), 4.3-4.6 (2H, m) 4.0-4.3 (2H, m), 3.2 (3H, s) | 287 (M ⁺ - CF ₃ SO ₃ , 100), 286 (30), 285 (27), 271 (17), 230 (10), 202 (11), 143 (29), 115 (7), 77 (10) |

[a] Solvent, DMSO-d₆ for **2a** and **9**; deuteriochloroform for **8c**, **10** and **11**; deuteriochloroform-trifluoroacetic acid for **2b**, **2c**, **3**, **4**, **5**, **8a** and **8b**.

To a solution of **4** (3 mmoles) in dry dichloromethane (50 ml) methyl trifluoromethanesulphonate (3 mmoles) was added. The reaction mixture was stirred under reflux temperature for 3 hours. After cooling to room temperature the precipitated solid was separated by filtration and recrystallized from the appropriate solvent to give **5** as colourless prisms (Table I).

N-Substituted-4,6-diphenylpyridine-2-thiones **6**. General Procedure.

To a solution of 4,6-diphenyl-2*H*-pyran-2-thione (10 mmoles) in boiling methanol (50 ml), the appropriate β-functionalized amine (12 mmoles) was added and the resulting mixture was heated at reflux temperature for 5 hours. After cooling, volatiles were removed under reduced pressure and the residual material was thoroughly washed with water, dried under vacuum and the crude pyridine-2-thione **6** was used without further purification in the next step.

Bicyclic Salts **8** and **9**. General Procedure.

To a solution of the appropriate pyridine-2-thione **6** (10 mmoles) in dry benzene (60 ml), methyl iodide (12 mmoles) was added and the reaction mixture was stirred at room temperature for 24 hours. The yellow precipitated solid was collected by filtration, washed with ether, dried and recrystallized from the appropriate solvent (Table I) to give the salt **8** or **9** as crystalline solid.

Compound **8c** was isolated as tetrafluoroborate from the iodide by anion exchange with tetrafluoroboric acid in ethanolic solution at reflux temperature for 1 hour.

5,7-Diphenyl-2,3-dihydroimidazo[1,2-*a*]pyridine (**10**).

A solution of 5,7-diphenyl-2,3-dihydro-1*H*-imidazo[1,2-*a*]pyridinium iodide (**9**) (5 mmoles) and potassium hydroxide (10 mmoles) in methanol (70 ml) was heated under reflux temperature for 10 hours. After cooling the solvent was removed under reduced pressure and the residual material was dissolved in dichloromethane (50 ml). The resulting solution was washed with water (2 \times 30 ml) and dried over anhydrous sodium sulfate. Elimination of the solvent under reduced pressure leads to the crude product which recrystallized from ethanol/ether yielding 5,7-diphenyl-2,3-dihydroimidazo[1,2-*a*]pyridine (**10**) as crystalline solid (Table I).

1-Methyl-5,7-diphenyl-2,3-dihydroimidazo[1,2-*a*]pyridinium Trifluoromethanesulphonate (**11**).

To a solution of 5,7-diphenyl-2,3-dihydroimidazo[1,2-*a*]pyridine (**10**) (10 mmoles) in dry benzene (50 ml), methyl trifluoromethanesulphonate (12 mmoles) in the same solvent (15 ml) was added dropwise with stirring at room temperature. After a short period of time a yellow-greenish solid was gradually separated out, which filtered, dried and recrystallized from ethanol gave **11** as crystalline solid (Table I).

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