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# PYRROLOBENZOXAZEPINE RING CONSTRUCTION THROUGH METALATION AND ELECTROPHILIC SUBSTITUTION OF *N*-(2-HYDROXYMETHYLPHENYL)PYRROLE

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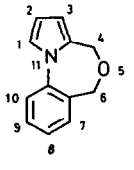
**Summary :** *N*-(2-Hydroxymethylphenyl)pyrrole was found to be amenable to selective α-metalation. Trapping with a variety of electrophilic reagents afforded the expected products with moderate to high yields. Derivatives formed by treatment of the organometallic intermediate with aldehydes, ketones or carbon dioxide could be cyclized to give pyrrolobenzoxazepines.

Like many other benzo annelated two-ring heterocycles [1], the 4*H*,6*H*-pyrrolo[1,2-*a*][4,1]benzoxazepine core (1) may be considered as a "pharmacophore". Members of the isomeric family of 5*H*,11*H*-pyrrolo[2,1-*c*][1,4]benzoxazepines (2) exhibit antinociceptive (hence analgesic) and central nervous system depressive activity [1, 2]. The structurally related imidazo[1,5-*a*][1,4]benzodiazepines, in particular Flumazenil (3), are competitive antagonists of the benzodiazepine (*e.g.*, diazepam) receptor [3].



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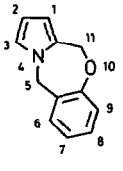

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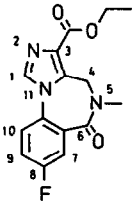

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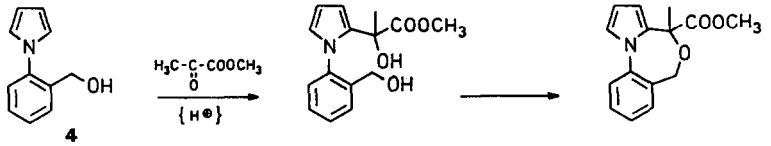
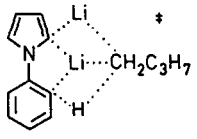
The standard preparation of the 4*H*,6*H*-pyrrolo[1,2-*a*][4,1]benzoxazepine ring system is based on the acid catalyzed attachment of a reactive carbonyl compound, for example ethyl pyruvate, to the α-position of *N*-(hydroxymethylphenyl)pyrrole (4) followed by cyclization. The reported over-all yields are poor (< 30%).

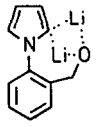


4

It should be possible to carry out such condensation reactions more efficiently and to extend its scope to less electrophilic carbonyl components if the pyrrole precursor could be metalated at the α-position. This would immensely and selectively increase the nucleophilicity of the α-carbon site. In a previous study [4] we have identified reaction conditions that allow to carry out a nearly quantitative α-monometalation of *N*-phenylpyrrole. Crucial for the success was to avoid the accumulation of *o*,α-dimetalated species the formation of which is kinetically favored by the neighboring group assistance (transition state **5**; for clarity, solvation and complexation being neglected) of the C(α),Li bond generated in the first stage. This complication should be attenuated with *N*-(2-hydroxymethylphenyl)pyrrole as the substrate since the newly formed α-carbon,metal bond would probably become associated with the alcoholate moiety (forming the intramolecular "mixed aggregate" **6** or oligomers thereof; M = metal) and thus be kept away from the unsubstituted *ortho* position.

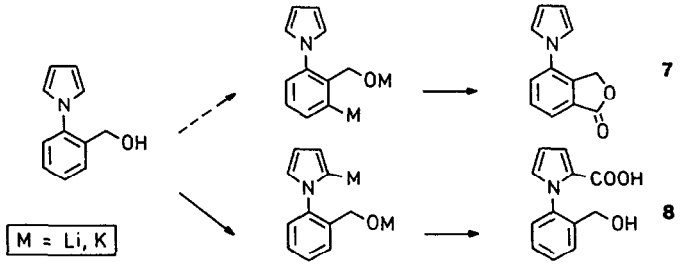



**5**



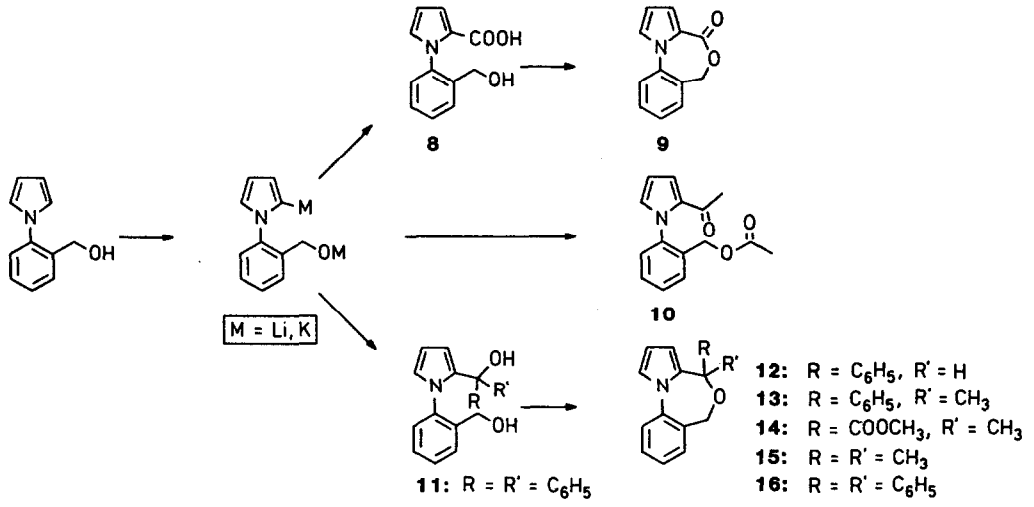

**6**

The α-metalation could be brought about very smoothly indeed with butyllithium activated by potassium *tert*-butoxide [4] as the base. After 2 h of metalation at -75 °C, carboxylation and neutralization, 83% of the acid **8** was isolated. On the other hand, with butyllithium activated by *N*,*N*,*N'*,*N'*-tetramethylethylenediamine (TMEDA) in diethyl ether (24 h at 0 °C) or in hexane (3 h at 65 °C), 2 : 1 or, respectively, 5 : 1 mixtures of the acid **8** and the lactone **7** were obtained, the latter obviously resulting from metalation at the *meta* position of the phenyl ring.

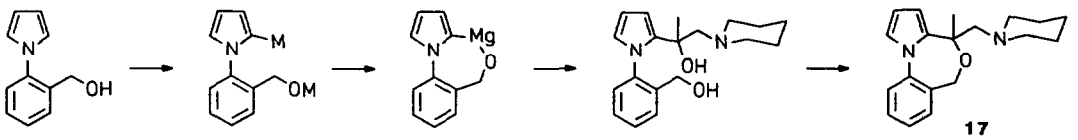



The α-deprotonated intermediate could be also trapped with a variety of different electrophiles such as acetic anhydride (giving 54% of the oxoacetate **10**) and aldehydes or ketones (giving, *e.g.*, 52% of diol **11**). The hydroxyacid **8** was cleanly converted into lactone **9** (76%) by treatment with *p*-toluenesulfonic acid in the presence of pyridine. The dehydration and cyclization of *N*-(2-hydroxymethylphenyl)-2-(α-hydroxyalkyl)pyrroles was most conveniently accomplished by adding silica gel and toluene to these diols before heating the mixture

3 h to 65 °C. In this way, the pyrrolobenzoxazepines **12** (67%), **13** (34%), **14** (31%), **15** (37%) and **16** (60%) were prepared without requiring isolation of their diol precursors. The parent ring system, pyrrolo[1,2-*a*][4,1]-benzoxazepine (R, R' = H) has already been reported previously [5].




This reaction sequence was finally applied to an amino substituted derivative. *N*-(2-Hydroxymethylphenyl)pyrrole (**4**) was consecutively submitted to superbase metalation [6] (M = K or Li), metal exchange with magnesium dibromide, addition of *N*-acetonylpiperidine and dehydration with silica gel. Pure, crystalline 4-methyl-4-(*N*-piperidylmethyl)-4*H*,6*H*-pyrrolo[1,2-*a*][4,1]benzoxazepine (**17**) was obtained with a 48% over-all yield.




## EXPERIMENTAL PART

### 1. Generalities

See also a related article [4].

*Starting materials* were purchased from Fluka AG (Buchs), unless literature sources or details of the preparation are given. *Butyllithium* and *potassium tert-butoxide* were supplied by CheMetall, Frankfurt, and Hüls, Troisdorf. All commercial reagents were used without further purification.

*Air and moisture sensitive compounds* were stored in Schlenk tubes or Schlenk burettes. They were protected by and handled under an atmosphere of 99.995% pure nitrogen.

Unless otherwise stated, *nuclear magnetic resonance spectra* of hydrogen-1 and carbon-13 nuclei were recorded of deuterochloroform solutions at 250 and 90.6 MHz, respectively. Chemical shifts refer to the signal of tetramethylsilane ($\delta$ = 0 ppm). Coupling constants ($J$) are measured in Hz. Abbreviations of coupling patterns: s (singlet), d (doublet), t (triplet), q (quadruplet), td (triplet of doublets) and m (multiplet).

*Mass spectra* were obtained at a 70 eV ionization potential. Whenever no molecular peak was observed under standard conditions, chemical ionization ("c.i.") in an ammonia atmosphere was applied. - *Elementary analyses* were performed by the laboratory of I. Beetz, D-96317 Kronach.

## 2. Metalation With Butyllithium Activated By *N,N,N',N'*-Tetramethylethylenediamine

At 0 °C, 1-(2-hydroxymethylphenyl)pyrrole [7] (4.3 g, 25 mol) was added to a solution of butyllithium (50 mmol) in hexane (30 mL) and diethyl ether (60 mL). After 24 h at 0 °C, the mixture was poured on an excess of freshly crushed dry ice. The product was extracted with water (2 × 50 mL) and the organic layer was discarded. An aliquot of the aqueous phase was neutralized and treated with diazomethane. By comparison with authentic samples and an "internal standard" (tetradecane), gas chromatography (2 m, 10% Apiezon-L, 230 °C) revealed the presence of lactone 7 (36%) and of methyl 1-(2-hydroxymethylphenyl)-2-pyrrylcarboxylate (60%), the ester derived from acid 8. The remainder of the alkaline phase was acidified to pH 1 and extracted with diethyl ether (3 × 25 mL). The combined organic layers were evaporated and the residue was crystallized from hexane and recrystallized from ethyl acetate to yield the acid 8 (mp 119 - 120 °C, dec.; see Section 3.a). The combined mother liquors were evaporated and the residue was repetitively crystallized from isopropyl alcohol to get pure *4-pyrrolo-1H,3H-isobenzofuran-1-one* (7) : mp 98 - 99 °C. - $^1$H-NMR : $\delta$ 7.9 (1 H, m), 7.7 (2 H, m), 6.99 (2 H, t-like m, $J$ ~ 2.1), 6.42 (2 H, t-like m, $J$ ~ 2.1), 5.45 (2 H, s). - $^{13}$C-NMR : $\delta$ 170.2 (1 C, s), 138.0 (1 C, s), 136.2 (1 C, s), 130.7 (1 C, d, $J$ 165), 127.9 (1 C, s), 127.2 (1 C, dd, $J$ 162, 7), 123.3 (1 C, d, $J$ 170), 120.0 (2 C, d, $J$ 185), 111.4 (2 C, d, $J$ 172), 68.8 (1 C, t, $J$ 154). - MS (c.i.) : 200 (30%, $M^+$ +1), 199 (100%, $M^+$), 143 (42%). - Analysis : calc. for $C_{12}H_9NO_2$ (199.21) C 72.35, H 4.55; found C 72.22, H 4.91%.

## 3. Metalation With Butyllithium Activated By Potassium *tert*-Butoxide

Precooled (-75 °C) tetrahydrofuran (0.10 L), potassium *tert*-butoxide (5.6 g, 50 mmol) and 1-(2-hydroxymethylphenyl)pyrrole [7] (4.3 g, 25 mmol) were consecutively added to butyllithium (50 mmol) from which the original solvent (hexane) had before been stripped off. After 2 h of vigorous stirring at -75 °C, the reaction mixture was treated with the electrophilic reagent. The product was isolated by extraction and purified by crystallization.

a) **1-(2-Hydroxymethylphenyl)pyrrole-2-carboxylic acid (8)** : 83%; mp 119 - 120 °C (dec.; recryst. from ethyl acetate). - $^1$H-NMR ($CD_3OD$) : $\delta$ 7.59 (1 H, dq, $J$ 7.5, 1.0), 7.44 (1 H, td, $J$ 7.5, 1.5), 7.32 (1 H, td, $J$ 7.6, 1.5), 7.14 (1 H, dd, $J$ 8.1, 1.5), 7.07 (1 H, dd, $J$ 3.9, 1.8), 6.90 (1 H, dd, $J$ 2.4, 1.8), 6.31 (1 H, dd, $J$ 4.0, 2.5), 4.38 (1 H, d, $J$ 13.5), 4.20 (1 H, d, $J$ 13.5). - MS (c.i.) : 218 (4%, $M^+$ +1), 217 (39%, $M^+$), 172 (100%), 154 (59%). - Analysis : calc. for $C_{12}H_{11}NO_3$ (217.22) C 66.35, H 5.10; found C 66.14, H 5.21%.

Treatment of acid 8 (10 mmol) in diethyl ether (20 mL) during 15 h at 25 °C with *p*-toluenesulfonyl chloride (10 mmol) and pyridine (15 mL) converted it to **4*H*,6*H*-pyrrolo[1,2-*a*][4,1]benzoxazepin-4-one (9)**; mp 99 - 101 °C (dec.; recryst. from ethyl acetate). - $^1$H-NMR : $\delta$ 7.4 (6 H, m), 6.46 (1 H, dd, $J$ 3.5, 2.6), 5.09 (2 H, s, broad). - $^{13}$C-NMR : $\delta$ 162.1 (1 C, s), 139.0 (1 C, s), 130.7 (1 C, dd, $J$ 163, 8), 129.8 (1 C, d, $J$ 161), 128.3 (1 C, s), 126.8 (1 C, dd, $J$ 163, 8), 125.3 (1 C, d, $J$ 187), 124.7 (1 C, s), 122.2 (1 C, d, $J$ 177), 121.6 (1 C, dd, $J$ 162, 8), 111.5 (1 C, d, $J$ 171), 67.3 (1 C, td, $J$ 149, 5). - MS (c.i.) : 201 (6%, $M^+$ + 2), 200 (33%, $M^+$ + 1), 199 (63%, $M^+$), 171 (27%), 143 (100%), 115 (40%). - Analysis : calc. for $C_{12}H_9NO_2$ (199.21) C 72.35, H 4.55; found C 72.27, H 4.51%.

b) **1-(2-Acetoxymethylphenyl)-2-acetylpyrrole (10)** : With acetic anhydride (4.7 mL, 5.1 g, 50 mmol); 54%; mp 58 - 59 °C (dec.; recryst. from hexane). - $^1$H-NMR : $\delta$ 7.48 (1 H, dd, $J$ 7.7, 1.9), 7.44 (1 H, td, $J$ 7.4, 1.4), 7.38 (1 H, td, $J$ 7.6, 1.9), 7.18 (1 H, dd, $J$ 7.3, 1.4), 7.07 (1 H, dd, $J$ 3.9, 1.6), 6.87 (1 H, dd, $J$ 2.4, 1.5), 6.32 (1 H, dd, $J$ 5.6, 2.1), 4.78 (2 H, s), 2.39 (3 H, s), 2.00 (3 H, s). - MS (c.i.) : 258 (3%, $M^+$ +1), 257 (10%, $M^+$), 197 (51%), 154 (100%). - Analysis : calc. for $C_{15}H_{15}NO_3$ (257.29) C 70.02, H 5.88; found C 69.85, H 5.81%.

c) **1-(Hydroxymethylphenyl)-2-(α,α-diphenylhydroxymethyl)pyrrole (11)** : With benzophenone (4.6 g, 25 mmol); 52%; mp 160 - 162 °C (dec.; recryst. from ethyl acetate). - $^1$H-NMR : 7.35 (1 H, td, *J* 7.7, 1.8), 7.2 (9 H, m), 7.0 (2 H, m), 6.90 (1 H, td, *J* 7.7, 1.6), 6.55 (1 H, dd, *J* 2.8, 1.8), 6.35 (1 H, dd, *J* 7.8, 1.0), 6.12 (1 H, dd, *J* 3.5, 2.8), 5.60 (1 H, dd, *J* 3.5, 1.8), 4.25 (2 H, d, *J* 5.9), 3.82 (1 H, s, broad), 3.44 (1 H, symm. m, q-like). - MS (c.i.) : 355 (6%, $M^+$), 260 (12%), 105 (100%). - Analysis : calc. for $C_{24}H_{21}NO_2$ (355.42) C 81.10, H 5.96; found C 81.34, H 6.01%.

In general, the products formed by reaction of the organometallic intermediate with electrophiles were not isolated as such. After neutralization and extraction, the crude reaction mixtures were evaporated and redissolved in warm toluene (0.10 L) to which silica gel (30 g) was added. After 3 h of stirring at 65 °C, the suspension was evaporated to dryness and the resulting powder was poured on top of a column filled with fresh silica gel (70 g) and hexane. Elution with a 1 : 4 (v/v) mixture of ethyl acetate and hexane gave the cyclized product.

d) **4-Phenyl-4*H*,6*H*-pyrrolo[1,2-*a*][4,1]benzoxazepine (12)** : With benzaldehyde (2.5 mL, 2.7 g, 25 mmol); 67%; mp 70 - 71 °C (recryst. from hexane). - $^1$H-NMR : δ 7.5 (5 H, m), 7.3 (4 H, m), 7.08 (1 H, dd, *J* 2.8, 1.7), 6.23 (1 H, td, *J* 3.2, 0.5), 5.69 (1 H, ddd, *J* 3.5, 1.4, 0.6), 5.47 (1 H, s), 4.69 (1 H, d, *J* 11.8), 4.67 (1 H, d, *J* 11.8). - $^{13}$C-NMR : δ 140.4 (1 C, s), 139.5 (1 C, s), 133.7 (1 C, s), 130.6 (1 C, d, *J* 164), 129.9 (1 C, s), 129.9 (1 C, d, *J* 162), 128.0 (2 C, d, *J* 159), 127.7 (1 C, d, *J* 160), 127.3 (1 C, d, *J* 160), 126.5 (2 C, dd, *J* 162, 7), 121.1 (1 C, d, *J* 186), 120.8 (1 C, d, *J* 161), 110.9 (1 C, d, *J* 171), 109.2 (1 C, d, *J* 172), 72.7 (1 C, d, *J* 144), 67.2 (1 C, td, *J* 145, 5). - MS (c.i.) : 262 (36%, $M^+$ +1), 261 (100%, $M^+$), 232 (59%). - Analysis : calc. for $C_{18}H_{15}NO$ (261.32), C 82.73, H 5.79; found C 82.87, H 5.80%.

e) **4-Methyl-4-phenyl-4*H*,6*H*-pyrrolo[1,2-*a*][4,1]benzoxazepine (13)** : Prior to the addition of acetophenone (2.9 mL, 3.0 g, 25 mmol), the organometallic intermediate was treated with magnesium dibromide etherate [8] (55 mmol); 34%; mp 99 - 101 °C (recryst. from isopropyl alcohol). - $^1$H-NMR : δ 7.21 (1 H, dd, *J* 7.2, 1.8), 6.9 (9 H, m), 6.54 (1 H, dd, *J* 3.7, 1.8), 6.36 (1 H, dd, *J* 3.5, 2.9), 4.67 (1 H, d, *J* 12.1), 4.63 (1 H, d, *J* 12.1), 1.78 (3 H, s). - $^{13}$C-NMR : 147.8 (1 C, s), 140.6 (1 C, s), 135.2 (1 C, s), 131.1 (1 C, s), 129.5 (1 C, dd, *J* 160, 7), 129.2 (1 C, dd, *J* 162, 9), 127.1 (2 C, dd, *J* 159, 7), 125.8 (1 C, d, *J* 160), 125.7 (1 C, d, *J* 160), 125.1 (2 C, dt, *J* 159, 7), 121.7 (1 C, dt, *J* 185, 8), 120.5 (1 C, dd, *J* 161, 7), 110.8 (1 C, d, *J* 171), 108.6 (1 C, d, *J* 172), 77.7 (1 C, s), 67.2 (1 C, td, *J* 145, 5), 31.6 (1 C, q, *J* 129). - MS (c.i.) : 276 (31%, $M^+$ +1), 275 (28%, $M^+$), 260 (74%), 105 (100%), 77 (31%). - Analysis: calc. for $C_{19}H_{17}NO$ (275.35) C 82.88, H 6.22; found C 82.65, H 6.33%.

f) **4-Methoxycarbonyl-4-methyl-4*H*,6*H*-pyrrolo[1,2-*a*][4,1]benzoxazepine (14)** : With methyl pyruvate (2.6 mL, 3.1 g, 30 mmol); 31%; mp 94 - 95 °C (recryst. from isopropyl alcohol). - $^1$H-NMR : δ 7.4 (4 H, m), 7.05 (1 H, dd, *J* 2.9, 1.7), 6.45 (1 H, dd, *J* 3.6, 1.6), 6.37 (1 H, dd, *J* 3.5, 2.9), 4.65 (1 H, d, *J* 12.0), 4.49 (1 H, d, *J* 12.0), 3.22 (3 H, s), 1.81 (3 H, s). - $^{13}$C-NMR : 174.6 (1 C, s), 140.0 (1 C, s), 131.6 (1 C, s) 130.6 (1 C, d, *J* 161), 131.1 (1 C, s), 130.0 (1 C, dd, *J* 162, 8), 126.7 (1 C, dd, *J* 162, 10), 121.3 (1 C, d, *J* 164), 121.3 (1 C, d, *J* 194), 109.9 (1 C, d, *J* 170), 109.3 (1 C, d, *J* 172), 75.7 (1 C, s), 67.0 (1 C, t, *J* 145), 52.2 (1 C, q, *J* 148), 24.6 (1 C, q, *J* 131). - MS (c.i.) : 258 (38%, $M^+$ +1), 257 (9%, $M^+$), 198 (100%). - Analysis: calc. for $C_{15}H_{15}NO_3$ (257.30) C 70.02, H 5.88; found C 71.03, H 6.02%.

g) **4,4-Dimethyl-4*H*,6*H*-pyrrolo[1,2-*a*][4,1]benzoxazepine (15)** : Prior to the addition of acetone (2.2 mL, 1.7 g, 30 mmol), the organometallic intermediate was treated with magnesium dibromide etherate (55 mmol); 37%; mp 86 - 87 °C (recryst. from isopropyl alcohol). - $^1$H-NMR : 7.4 (2 H, m), 7.3 (1 H, m), 7.2 (1 H, m), 7.01 (1 H, t, *J* 2.3), 6.27 (2 H, symm. m), 4.47 (2 H, s), 1.37 (6 H, s). - $^{13}$C-NMR : δ 140.9 (1 C, s), 136.6 (1 C, s), 132.1 (1 C, s), 129.6 (1 C, d, *J* 162), 129.5 (1 C, d, *J* 162), 126.0 (1 C, d, *J* 163), 122.3 (1 C, dt, *J* 194, 8), 120.4 (1 C, dd, *J* 160, 8), 108.7 (1 C, d, *J* 170), 108.6 (1 C, d, *J* 170), 74.6 (1 C, s), 66.7 (1 C, td, *J* 142, 4), 30.1 (2 C, q, *J* 125). - MS (c.i.) : 214 (12%, $M^+$ +1), 213 (36%, $M^+$), 198 (100%). - Analysis: calc. for $C_{14}H_{15}NO$ (213.28) C 78.84, H 7.09; found C 79.28, H 7.17%.

h) **4,4-Diphenyl-4*H*,6*H*-pyrrolo[1,2-*a*][4,1]benzoxazepine (16)** : With benzophenone (5.5 g, 30 mmol); 60%; mp 152 - 154 °C (dec.; recryst. from ethyl acetate). - $^1$H-NMR : δ 7.1 (15 H, m), 6.26 (1 H, t, *J* 3.2), 5.65 (1 H, dd,

*J* 3.5, 1.5), 4.93 (2 H, s). - $^{13}$C-NMR : $\delta$ 145.4 (2 C, s), 140.8 (1 C, s) 135.4 (1 C, s), 130.9 (1 C, s), 129.5 (1 C, d, *J* 160), 129.3 (1 C, d, *J* 162), 127.6 (2 C, d, *J* 160), 127.4 (2 C, d, *J* 160), 127.3 (2 C, d *J* 160), 127.2 (2 C, d, *J* 160), 127.0 (1 C, d, *J* 167), 126.8 (1 C, d, *J* 167), 125.7 (1 C, d, *J* 162), 122.1 (1 C, dt, *J* 185, 8), 120.5 (1 C, dd, *J* 161, 8), 111.3 (1 C, *J* 171), 108.3 (1 C, d, *J* 172), 82.8 (1 C, s), 67.4 (1 C, td, *J* 145, 5). - MS (c.i.) : 338 (15%, $M^+$ + 1), 337 (49%, $M^+$), 260 (66%), 105 (100%). - Analysis: calc. for $C_{24}H_{19}NO$ (337.42) C 85.43, H 5.68; found C 85.27, H 5.75%.

i) **4-Methyl-4-(1-piperidylmethyl)-4*H*,6*H*-pyrrolo[1,2-*a*][4,1]benzoxazepine** (17) : Prior to the addition of 1-piperidylacetone [9] (3.5 g, 25 mmol), the organolithium intermediate was treated with magnesium dibromide etherate (55 mmol); 48%; mp 57 - 58 °C (recryst. from hexane). - $^1$H-NMR : $\delta$ 7.4 (2 H, m), 7.31 (1 H, d, broad, *J* 7.7), 7.2 (1 H, m), 6.99 (1 H, dd, *J* 2.8, 2.0), 6.29 (2 H, symm. m), 4.59 (1 H, d, *J* 12.3), 4.42 (1 H, d, *J* 12.3), 2.13 (4 H, symm. m), 1.90 (2 H, ddd, *J* 12.0, 7.0, 4.3), 1.61 (3 H, s), 1.3 (6 H, m). - MS : 296 (12%, $M^+$), 198 (48%), 155 (81%), 98 (100%). - Analysis : calc. for $C_{19}H_{24}N_2O$ (296.41) C 76.99, H 8.16; found C 76.91, H 8.17%.

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## REFERENCES

[1] R.C. Effland, L. Davis, *J. Heterocycl. Chem.* **22** (1985), 1071.
[2] K.J. Kapples, R.C. Effland, *J. Heterocycl. Chem.* **30** (1993), 177.
[3] W. Hunkeler, H. Möhler, L. Pieri, P. Polc, E.P. Bonetti, R. Cumin, R. Schaffner, W. Haefely, Nature **290** (1981), 514.
[4] F. Faigl, M. Schlosser, "*N*-Phenylpyrrole : A Kinetic, Though Not Thermodynamic Preference for Dilithiation", *Tetrahedron*, in press (1993).
[5] S. Massa, F. Corelli, G. Stefancich, *J. Heterocycl. Chem.* **18** (1981), 829; G.W.H. Cheeseman, S.A. Eccleshall, T. Thornton, *J. Heterocycl. Chem.* **22** (1985), 809.
[6] M. Schlosser, *J. Organomet. Chem.* **8** (1967), 9; *Pure Appl. Chem.* **60** (1988), 1627.
[7] F.Z. Basha, R.W. Franck, *J. Org. Chem.* **43** (1978), 3415.
[8] L. Tissier, V. Grignard, *C.R. Séances Acad. Sci.* **132** (1901), 895.
[9] R. Stoermer, O. Burkert, *Ber. Dtsch. Chem. Ges.* **28** (1985), 1250.

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