

Synthesis of 6-(arylmethylidene)octahydropyrrolo[1,2-*a*]pyrimidines and 5-(arylmethylidene)hexahydropyrrolo[1,2-*a*]imidazoles by the interaction of alk-4-ynals with α,ω -diaminoalkanes in DMSO in the presence of KOH

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(*E*)-6-Benzilydeneoctahydropyrrolo[1,2-*a*]pyrimidine **5d** was prepared from aldehyde **1b** and 1,3-diaminopropane **2b** and isolated in 42% yield by recrystallization from hexane, mp 68–71°C. ¹H NMR, δ : 1.48–1.78 (m, 3H, NCH₂CH₂CH₂N, =CCH₂CHH), 2.03 (br.s, 1H, NH), 2.22 (dddd, 1H, =CCH₂CHH, ²*J* 11.6 Hz, ³*J* 8.7 Hz, ³*J* 5.8 Hz, ³*J* 2.9 Hz), 2.55–3.03 (m, 4H, =CCH₂, NCHHCH₂CHHNH), 3.21 (br.d, 1H, NCH₂CH₂CHHNH, *J* 13.5 Hz), 3.70 (br.d, 1H, NCHHCH₂CH₂NH, *J* 12.2 Hz), 3.87 (dd, 1H, NCHN, *J* 8.1 Hz, *J* 5.8 Hz), 5.21 (br.s, 1H, PhCH=), 6.99 (br.t, 1H, Ph, *J* 6.8 Hz), 7.15–7.30 (m, 4H, Ph). ¹³C NMR, δ : 24.8, 27.2, 28.6 (C³, cyclo-C₄N₂, C⁷, C⁸, cyclo-C₄N), 43.3, 44.8 (C², C⁴, cyclo-C₄N₂), 75.3 (NCHN), 93.3 (PhCH=), 122.7, 126.0, 128.1 (Ph), 139.6 (C¹, Ph), 147.5 (PhCH=C). MS, *m/z*: 214 [M⁺], 213 [M – H]⁺. Found (%): C, 78.65; H, 8.23; N, 13.36. Calc. for C₁₄H₁₈N₂ (%): C, 78.46; H, 8.47; N, 13.07.

(*E*)-6-Benzylidene-1-methyloctahydropyrrolo[1,2-*a*]pyrimidine **5e** was prepared from aldehyde **1b** and *N*-methyl-1,3-diaminopropane **2c** and isolated in 51% yield by recrystallization from hexane. ¹H NMR, δ : 1.60–1.74 (m, 1H, NCH₂CHHCH₂N), 1.76–2.23 (m, 4H, NCH₂CHHCH₂N, =CCH₂CH₂, MeNCHHCH₂CH₂N), 2.29 (s, 3H, NMe), 2.57–2.81 (m, 2H, =CCHH, MeNCHHCH₂CH₂N), 2.91–3.07 (m, 2H, =CCHH, MeNCH₂CH₂CHHN), 3.08 (dd, 1H, NCHN, *J* 7.5 Hz, *J* 5.9 Hz), 3.60–3.72 (m, 1H, MeNCH₂CH₂CHHN), 5.25 (br.s, 1H, PhCH=), 7.00 (br.t, 1H, Ph, *J* 6.8 Hz), 7.15–7.30 (m, 4H, Ph). ¹³C NMR, δ : 23.8, 27.5, 27.7 (C³, cyclo-C₄N₂, C⁷, C⁸, cyclo-C₄N), 41.5 (NMe), 42.7 (C⁴, cyclo-C₄N₂), 55.2 (C², cyclo-C₄N₂), 82.5 (NCHN), 93.8 (PhCH=), 122.9, 126.2, 128.2 (Ph), 139.8 (C¹, Ph), 148.2 (PhCH=C). MS, *m/z*: 228 [M⁺], 227 [M – H]⁺. Found (%): C, 78.56; H, 9.03; N, 12.01. Calc. for C₁₅H₂₀N₂ (%): C, 78.90; H, 8.83; N, 12.27.

(*E*)-6-(4-Fluorobenzylidene)-1,8,8-trimethyloctahydropyrrolo[1,2-*a*]pyrimidine **5f** was prepared from aldehyde **1c** and *N*-methyl-1,3-diaminopropane **2c** and isolated in 60% yield by recrystallization from hexane, mp 98–100 °C. ¹H NMR, δ : 1.12 (s, 3H, Me), 1.27 (s, 3H, Me), 1.60–1.74 (m, 1H, NCH₂CHHCH₂N), 1.78–2.00 (m, 1H, NCH₂CHHCH₂N), 2.04–2.21 (m, 1H, MeNCHHCH₂CH₂N), 2.32 (s, 3H, NMe), 2.46 (br.d, 1H, =CCHH, *J* 15.6 Hz), 2.51 (ddd, 1H,

MeNCHHCH₂CH₂N, ²J 12.0 Hz, ³J 12.0 Hz, ³J 4.0 Hz), 2.66 (br.d, 1H, =CCHH, J 15.6 Hz), 2.73 (s, 1H, NCHN), 2.94 (ddd, 1H, MeNCH₂CH₂CHHN, ²J 10.9 Hz, ³J 3.4 Hz, ³J 3.4 Hz), 3.50–3.62 (m, 1H, MeNCH₂CH₂CHHN), 5.15 (br.s, 1H, 4-FC₆H₄CH=), 6.85–7.15 (m, 4H, Ph). ¹³C NMR, δ: 22.5 (Me), 24.1 (C³, cyclo-C₄N₂), 28.1 (Me), 38.3 (C⁸, cyclo-C₄N), 42.3 (NMe), 43.0, 45.8 (C⁴, cyclo-C₄N₂, C⁷, cyclo-C₄N), 56.3 (C², cyclo-C₄N₂), 90.1, 92.0 (NCHN, 4-FC₆H₄CH=), 115.0 (d, C³, C⁵, Ph, J_{CF} 21 Hz), 127.4 (d, C², C⁶, Ph, J_{CF} 7.2 Hz), 136.0 (C¹, Ph, J_{CF} 2.7 Hz), 148.6 (4-FC₆H₄CH=C), 162.0 (d, C⁴, Ph, J_{CF} 242 Hz). MS, m/z: 274 [M⁺], 273 [M – H]⁺. Found (%): C, 74.71; H, 8.21 N, 9.98. Calc. for C₁₇H₂₃FN₂ (%): C, 74.42; H, 8.45; N, 10.21.

(E)-1-Methyl-6-(4-trifluoromethylbenzylidene)octahydropyrrolo[1,2-a]pyrimidine **5g** was prepared from aldehyde **1d** and N-methyl-1,3-diaminopropane **2c** and isolated in 45% yield by recrystallization from hexane, mp 109–110 °C. ¹H NMR, δ: 1.60–1.74 (m, 1H, NCH₂CHHCH₂N), 1.76–2.23 (m, 4H, NCH₂CHHCH₂N, =CCH₂CH₂, MeNCHHCH₂CH₂N), 2.26 (s, 3H, NMe), 2.57–2.80 (m, 2H, =CCHH, MeNCHHCH₂CH₂N), 2.88–3.08 (m, 2H, =CCHH, MeNCH₂CH₂CHHN), 3.16 (dd, 1H, NCHN, J 7.2 Hz, J 5.7 Hz), 3.60–3.73 (m, 1H, MeNCH₂CH₂CHHN), 5.20 (br.s, 1H, 4-F₃CC₆H₄CH=), 7.21 (br.d, 2H, Ph, J 8.3 Hz), 7.43 (br.d, 2H, Ph, J 8.3 Hz). ¹³C NMR, δ: 23.7, 27.4, 28.3 (C³, cyclo-C₄N₂, C⁷, C⁸, cyclo-C₄N), 41.4 (NMe), 42.5 (C⁴, cyclo-C₄N₂), 55.2 (C², cyclo-C₄N₂), 82.5 (NCHN), 93.8 (4-F₃CC₆H₄CH=), 123.9 (q, C⁴, Ph, J 32 Hz), 124.8 (q, CF₃, J_{CF} 270 Hz), 125.0 (q, C³, C⁵, Ph, J_{CF} 3.7 Hz), 125.6 (C², C⁶, Ph), 143.8 (C¹, Ph), 150.5 (4-F₃CC₆H₄CH=C). MS, m/z: 296 [M⁺], 295 [M – H]⁺. Found (%): C, 65.21; H, 6.21; N, 9.69. Calc. for C₁₆H₁₉F₃N₂ (%): C, 64.85; H, 6.46; N, 9.45.

(E)-1,8,8-Trimethyl-6-(2-thienylmethylidene)octahydropyrrolo[1,2-a]pyrimidine **5h** was prepared from aldehyde **1e** and N-methyl-1,3-diaminopropane **2c** and isolated in 58% yield by recrystallization from hexane, mp 100–102 °C. ¹H NMR, δ: 1.13 (s, 3H, Me), 1.30 (s, 3H, Me), 1.60–1.74 (m, 1H, NCH₂CHHCH₂N), 1.76–2.00 (m, 1H, NCH₂CHHCH₂N), 2.00–2.15 (m, 1H, MeNCHHCH₂CH₂N), 2.29 (s, 3H, NMe), 2.44 (br.d, 1H, =CCHH, J 16.5 Hz), 2.50 (ddd, 1H, MeNCHHCH₂CH₂N, ²J 12.0 Hz, ³J 12.0 Hz, ³J 3.9 Hz), 2.69 (br.d, 1H, =CCHH, J 16.5 Hz), 2.75 (s, 1H, NCHN), 2.93 (ddd, 1H, MeNCH₂CH₂CHHN, ²J 10.9 Hz, ³J 3.4 Hz, ³J 3.4 Hz), 3.50–3.62 (m, 1H, MeNCH₂CH₂CHHN), 5.48 (br.s, 1H, ThiCH=), 6.65 (dd, 1H, thienyl, J 3.4 Hz, J 1.4 Hz), 6.88–6.98 (m, 2H, thienyl). ¹³C NMR, δ: 22.6 (Me), 24.0 (C³, cyclo-C₄N₂), 28.1 (Me), 38.5 (C⁸, cyclo-C₄N), 42.3 (NMe); 42.9, 45.9 (C⁴, cyclo-C₄N₂, C⁷, cyclo-C₄N), 56.2 (C², cyclo-C₄N₂), 87.4, 90.3 (NCHN, ThiCH=), 119.4, 120.2, 127.0 (thienyl), 144.1 (C¹, thienyl), 146.2 (ThiCH=C). MS, m/z: 262 [M⁺], 261 [M – H]⁺. Found (%): C, 68.32; H, 8.69; N, 10.97. Calc. for C₁₅H₂₂N₂S (%): C, 68.65; H, 8.45; N, 10.68.